Development of high load constructed wetlands for treatment of wastewater

Autor/a: Ana Pascual Formoso

Tesis doctoral UDC / Año 2023

Directores:

Manuel Soto Castiñeira

Carlos Alberto Arias

Juan Antonio Álvarez Rodríguez

Tutor: Manuel Soto Castiñeira

Programa de doctorado en Ciencia y Tecnología Ambiental

UNIVERSIDADE DA CORUÑA

Agradecimientos

Esta tesis es el resultado de muchos años de trabajo y esfuerzo. La combinación de compromiso personal y profesional, junto con los desafíos y cambios en mi vida familiar, hacen de este logro una conquista aún más significativa. Este arduo proceso ha merecido la pena y para mi representa todo un logro en mi vida.

Con estas palabras me gustaría reflejar mi gratitud hacia las personas que me han respaldado y que han sido una fuente constante de inspiración y apoyo.

Es importante reconocer el papel fundamental que desempeñaron mis directores de tesis y colaboradores en la realización de este trabajo de investigación. Debo darle las gracias de manera muy especial y sincera a Manuel Soto por todo su apoyo, paciencia y su participación activa durante todo el desarrollo de esta tesis. Carlos A. Arias por su actitud positiva, por animarme a seguir adelante, creer en mí y sus inestimables correcciones, siempre acertadas. Juan Álvarez por su disponibilidad y su acompañamiento durante muchos años no solo como doctoranda sino como compañera.

Me gustaría darle las gracias a todos los coautores de los artículos científicos que se presentan en esta tesis por sus aportaciones y colaboraciones. Entre todos ellos tengo que hacer una especial mención para David de la Varga, un gran compañero y mejor persona, siempre dispuesto a ayudar y echar una mano.

A todos mis compañeros de departamento especialmente mi amiga Paula Villar compañera de muchas horas de laboratorio, de muestreo, de análisis y sobre todo de buenos momentos y recuerdos. Muchas gracias a Rocío Pena por su proactividad y su rapidez a la hora de resolución de problemas. También quiero darle las gracias a Santiago Cuervo por los últimos años trabajando codo con codo y por su "vamos Anita" con el que siempre salía una sonrisa.

A mis amigas y traductoras Mar Vázquez y Lucía Rodríguez. Gracias por tener el ánimo y la paciencia de leer mis artículos y por vuestra inestimable ayuda con el inglés.

Me gustaría agradecer a mis padres todo su trabajo y esfuerzo a lo largo de todos estos años. Ellos me han enseñado que cuando uno quiere conseguir un objetivo con trabajo, esfuerzo y sacrificio todo es posible. También quiero dar las gracias a mi hermano y mi cuñada por su apoyo y por siempre estar disponibles.

Finalmente, quiero darle las gracias de una manera muy especial a mi marido Philippe, por su amor, su confianza y por todo su apoyo. Gracias por estar a mi lado y por hacerme creer que todo es posible. Gracias por recordarme que el final de esta tesis estaba en mis manos y más cerca de lo que parecía. A mis hijos Marcos y Lara, por su amor, cariño y por sacar lo mejor de mí. Han sido fuente de inspiración y motivación. En ciertos momentos he estado a punto de tirar la toalla, pero el darles ejemplo ha sido la fuerza que me ha llevado a no rendirme y seguir adelante. Como dijo el filósofo George Santayana: "La familia es una de las obras maestras de la naturaleza" y yo tengo la suerte de que mi familia es el cuadro en el que está pintando mi universo.

Table of contents

List of tables

List of figures

Acronym list

Acronym	Full text
ARGs	Antibiotic resistance genes
BOD_5	5-day (carbonaceous) Biochemical Oxygen Demand
COD	Chemical Oxygen Demand
CODin	COD influent concentration
CWs	Constructed wetlands
ER	Effluent recirculation
FWS	Free water surface
GI	Green Infrastructure
HUSB	Hydrolitc upflow sludge blanket
HLR	Hydraulic loading rate
HF	Horizontal Flow
HRT	Hydraulic retention time
O&M	Operation and maintenance
Org. N	Organic nitrogen
TP	Total phosphorus
TN	Total nitrogen
TSS	Total suspended solids
SLR	Surface loading rate
SLR_{COD}	Surface loading rate for COD
SRRs	Surface removal rates
SSF	Subsurface flow
UASB	up-flow anaerobic sludge blanket
VF	Vertical flow
VLR_{COD}	volumetric loading rate of COD
VLR_{TSS}	Volumetric loading rate of TSS
WW	Wastewater

Abstract

Constructed wetlands (CWs) technology is an established green multi-purpose option for water management and wastewater (WW) treatment, with numerous effectively proven applications around the world and multiple environmental and economic advantages. Their adaptability and low operation and maintenance (O&M) requirements make them a sustainable and cost-effective choice for various WW treatment applications.

CWs have been widely applied for over 50 years, initially for municipal wastewater and later for industrial and agricultural wastewater, livestock farm effluent, landfill leachate, and stormwater runoff. Industrial wastewater often requires pre-treatment due to its distinct composition. The introduction of oxygen in CWs, known as aerated CWs, enhances treatment efficiency, especially for nitrification and denitrification processes. These systems can be operated intermittently to improve total nitrogen removal. Aeration strategies can vary in intensity, making aerated CWs flexible and effective in removing nitrogen and organic matter.

The primary objective of this thesis is to investigate and optimise various factors affecting WW treatment in aerated CWs, encompassing urban and industrial WW, with the aim of enhancing the design parameters and future implementation of these systems. Additionally, the thesis seeks to evaluate the feasibility of employing the hydrolitc upflow sludge blanket (HUSB) reactor followed by CWs configuration for treating diverse sources of WW, including urban, food industry, and winery WW, and to assess the impact of design and operational parameters on treatment efficiency.

Research findings are intended to enhance the understanding of guidelines for CWs design, operation, and maintenance. Being carried out at outdoors pilot and full scale systems, the study spans several years and focuses on crucial factors such as unit performance, phosphorus removal, HUSB reactor for pre-treatment to prevent clogging risks in CWs, the influence of bed depth in aerated CWs, and treatment efficiency.

This research holds significant relevance in improving the design of efficient and cost-effective aerated CWs systems. It addresses the need for a better understanding of the internal processes involved in these systems and seeks to provide valuable performance data and information to guide the design and operation of aerated CWs.

Resumen

La tecnología de humedales artificiales (CWs) es una opción ecológica polivalente para la gestión del agua y el tratamiento de aguas residuales, con numerosas aplicaciones probadas en todo el mundo y múltiples ventajas medioambientales y económicas. Su adaptabilidad y sus reducidos requisitos de operación y mantenimiento los convierten en una opción sostenible y rentable para diversas aplicaciones de tratamiento de aguas residuales.

Las plantas de tratamiento de aguas llevan más de 50 años aplicándose de forma generalizada, inicialmente a las aguas residuales municipales y más tarde a los efluentes industriales, las aguas residuales agrícolas, los efluentes de explotaciones ganaderas, los lixiviados de vertedero y las aguas pluviales de escorrentía. Las aguas residuales industriales suelen requerir pretratamiento debido a su distinta composición. La introducción de oxígeno en los CW, conocidos como CW aireados, mejora la eficacia del tratamiento, especialmente en los procesos de nitrificación y desnitrificación. Estos sistemas pueden funcionar de forma intermitente para mejorar la eliminación total de nitrógeno. Las estrategias de aireación pueden variar en intensidad, haciendo que los CWs aireados sean flexibles y efectivos en la eliminación de nitrógeno y materia orgánica.

El objetivo principal de esta tesis es investigar y optimizar diversos factores que afectan al tratamiento de aguas residuales en CWs aireadas, abarcando las aguas residuales urbanas e industriales, con el fin de mejorar los parámetros de diseño y la futura implementación de estos sistemas. Además, la tesis pretende evaluar la viabilidad de emplear la configuración híbrida de manta de lodos de flujo ascendente (HUSB) seguida de CWs para tratar diversas fuentes de WW, incluyendo WW urbanas, de la industria alimentaria y de bodegas, y evaluar el impacto de los parámetros de diseño y operación en la eficiencia del tratamiento.

Los resultados de la investigación pretenden mejorar la comprensión de las directrices para el diseño, funcionamiento y mantenimiento de los CW. El estudio abarca varios años y se centra en factores cruciales como el rendimiento de la unidad, la eliminación de fósforo, el reactor HUSB para el pretratamiento con el fin de prevenir los riesgos de obstrucción en las CW, la influencia de la profundidad del lecho en las CW aireadas y la eficiencia del tratamiento.

Esta investigación es de gran relevancia para mejorar el diseño de sistemas de CWs aireados eficientes y rentables. Aborda la necesidad de una mejor comprensión de los procesos internos implicados en estos sistemas y trata de proporcionar valiosos datos de rendimiento e información para guiar el diseño y el funcionamiento de los CW aireados.

Resumo

A tecnoloxía de humedais artificiais (CWs) é unha opción ecolóxica polivalente para a xestión da auga e o tratamento das augas residuais, con numerosas aplicacións probadas en todo o mundo e múltiples ventaxas medioambientais e económicas. A súa adaptabilidade e os seus reducidos requisitos de operación e mantemento convértenos nunha opción sostible e rendible para diversas aplicacións de tratamento de augas residuais.

As plantas de tratamento de augas levan máis de 50 anos aplicándose de forma xeneralizada, inicialmente ás augas residuais municipais e máis tarde aos efluentes industriais, as augas residuais agrícolas, os efluentes de explotacións gandeiras, os lixiviados de entulleira e as augas pluviais de escorrentía. As augas residuais industriais adoitan requirir pretratamiento debido á súa distinta composición. A introdución de osíxeno nas CW, coñecidas como CW aireadas, mellora a eficacia do tratamento, especialmente nos procesos de nitrificación e desnitrificación. Estes sistemas poden funcionar de forma intermitente para mellorar a eliminación total de nitróxeno. As estratexias de aireación poden variar en intensidade, facendo que os CWs aireados sexan flexibles e efectivos na eliminación de nitróxeno e materia orgánica.

O obxectivo principal desta tese é investigar e optimizar diversos factores que afectan o tratamento de augas residuais en CWs aireadas, abarcando as augas residuais urbanas e industriais, co fin de mellorar os parámetros de deseño e a futura implementación destes sistemas. Ademais, a tese pretende avaliar a viabilidade de empregar a configuración híbrida do reactor de manto de lodos de fluxo ascendente (HUSB) seguida de CWs para tratar diversas fontes de WW, incluíndo WW urbanas, da industria alimentaria e de adegas, e avaliar o impacto dos parámetros de deseño e operación na eficiencia do tratamento.

Os resultados da investigación pretenden mellorar a comprensión das directrices para o deseño, funcionamento e mantemento dos CW. O estudo abarca varios anos e céntrase en factores cruciais como o rendemento da unidade, a eliminación de fósforo, o reactor HUSB para o pretratamiento co fin de previr os riscos de obstrución nas CW, a influencia da profundidade do leito nas CW aireadas e a eficiencia do tratamento.

Esta investigación é de gran relevancia para mellorar o deseño de sistemas de CWs aireados eficientes e rendibles. Aborda a necesidade dunha mellor comprensión dos procesos internos implicados nestes sistemas e trata de proporcionar valiosos datos de rendemento e información para guiar o deseño e o funcionamento dos CW aireados.

1. Introduction

1.1 Conceptual framework

Green Infrastructure (GI), Natural Based Solutions or Ecological and Natural Infrastructure are often used to describe similar approaches to remediate pollution. These systems are becoming increasingly recognised as an important opportunity for addressing the complex challenges of waste management. GI approach refers to natural or systems that mimic natural processes that have the potential to mitigate pollution while providing services for water resources management with equivalent or similar benefits to conventional (built) "grey" water infrastructure (Štrbac et al., 2023).

GI is the "strategic use of networks of natural lands, working landscapes, and other open spaces to conserve ecosystem values and functions and provide associated benefits to human populations" (Allen, 2013).

Blue-green infrastructure is also a term, used interchangeably with GI, to describe things like rain gardens or reed beds that treat WW. GI is generally decentralised, meaning water is captured and treated where it falls, rather than being transported to a treatment facility (Andoh B., et al., 2014).

Grey infrastructure refers to human-engineered infrastructure for water resources, such as water and WW treatment plants, pipelines, and dams. Grey infrastructure typically refers to components of a centralised approach to water management (Dolman N., et al., 2021)

In many developed and developing countries, governments, and communities are under constant pressure to optimise and expand water management infrastructures to provide the ever growing demand for water, energy and food. Treatment of WW from small and disperse populations is one of the most important problems due to decentralised location, limited economic resources and lack of specialised personnel. Investments in GI have been identified as one of the main building blocks for a transition to a Green Economy. However, many GI remain relatively novel solutions, presenting important challenges and unknowns in terms of their (co)design, operation, maintenance and how to establish them properly (Dolman N., et al., 2021).

1.2 CWs: option for WW management

Constructed wetlands (CWs) are GI solutions, design to tackle water and sludge pollution to meet discharge standards. CWs are engineered systems designed to optimise processes found in natural environments and are therefore considered environmentally friendly and sustainable options for WW treatment. Compared to other WW treatment technologies, treatment wetlands have low operation and maintenance (O&M) requirements and are robust in that performance is less susceptible to input variations. Treatment wetlands can effectively treat raw, primary, secondary or tertiary treated sewage and many types of agricultural and industrial WW. The processes to achieve the treatment in CWs include sedimentation, sorption, precipitation, evapotranspiration, volatilisation, photodegradation, diffusion, plant uptake, and microbial degradation processes such as nitrification, denitrification, sulphate reduction, carbon metabolisation, among others (Dotro et al., 2017).

The two main types of CWs systems are free water surface (FWS) systems and subsurface flow (SSF) systems, although other types exist Figure 1. Here's a brief overview of these systems (Dotro et al., 2017):

1. **FWS Systems:**

- FWS systems are shallow basins, typically ranging from 0.3 to 1.0 meters in depth.
- In FWS systems, WW flows freely, exposed to the atmosphere, like it would in natural ponds or wetlands.
- Aquatic vegetation is often planted in these systems, and it may be rooted in the bottom or floating plants such as water hyacinths, water lentil and water lettuce. water lilies.
- The vegetation helps in the treatment process by absorbing nutrients and providing habitat for microorganisms that break down pollutants.
- FWS systems are effective at removing pollutants through physical, chemical, and biological processes.

2. **SSF Systems:**

- SSF systems are designed as planted beds filled with porous materials like sand, gravel and others.
- WW is applied at the beginning of the bed and percolates through the porous media, allowing treatment to occur beneath the surface.
- Depending on the direction of flow through the media, there are two main types of SSF systems:
    - Horizontal Flow (HF) Systems: In HF systems, WW flows horizontally through the porous media from one end of the bed to the other.
    - Vertical Flow (VF) Systems: In VF systems, WW flows vertically downward through the porous media.
- In SSF systems, microorganisms attached to the porous media help break down organic matter and remove pollutants.
- SSF systems are particularly effective at treating organic matter and nutrients in WW.

Both FWS and SSF systems are sustainable and cost-effective alternatives for treating various types of WW, such as domestic sewage or industrial effluents. The choice between these systems depends on the specific treatment goals, site conditions, and regulatory requirements (Vymazal, 2010).

Subsurface flow CWs are subdivided into HF and VF wetlands depending on the direction of water flow (Figure 1, Table 1). In order to prevent clogging of the porous filter material, HF and VF wetlands are generally used for secondary treatment of WW. VF wetlands for treating screened raw WW have also been introduced and successfully applied (Kadlec and Wallace, 2009).

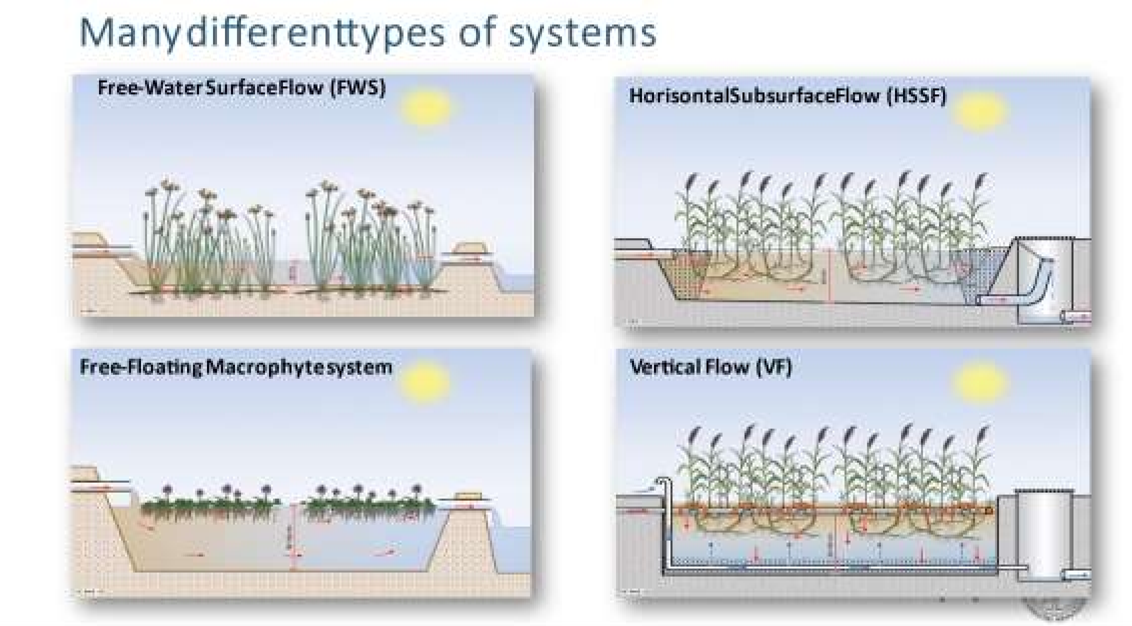


Figure 1: Overview schematics of treatment wetlands. Top Vertical Flow Treatment Wetland, bottom Horizontal Flow Treatment Wetland.

Table 1: Presents a summary of the two main treatment wetlands types covered in this thesis.

Type	Short description
HF treatment wetland	• WW flows horizontally through a sand or gravel filled filter whereby the water level is kept below the surface. • Due to the water-saturated condition mainly anaerobic degradation processes occur. • Effective primary treatment is required to remove particulate matter to prevent clogging of the filter. • Plants (macrophytes) are used. • They are used for secondary or tertiary treatment.
VF treatment wetland	• WW is intermittently loaded on the surface of the filter and percolates vertically through the filter.

	• Between two loadings air re-enters the pores and aerates the filter so that mainly aerobic degradation processes occur. • Effective primary treatment is required to remove particulate matter to prevent clogging of the filter. • Macrophytes are used.

The key difference between HF and VF wetlands lies in how they are designed to handle water and the oxygen levels within the system, which will determine the process for occurring in the system. Note that here we refer as VF only to intermittently fed and drained VF systems, in such a way that they create an unsaturated medium, since VF systems could also be operated with a saturated medium. VF systems, with their intermittent loading and passive aeration, provide enhanced oxygenation and can better support aerobic biological processes, making them more effective at treating certain pollutants, particularly organic matter and ammonium (NH_4^+). HF systems, on the other hand, are water saturated and may have a combination of aerobic and anaerobic conditions, which can be suitable for different types of WW treatment depending on the desired outcomes and specific site conditions e.g denitrification HF are usually operated in conditions of permanent water saturation which limits oxygen transfer and therefore nitrification only occurs at a low rate (Dotro et al., 2017).

Hybrid CWs systems are a combination of HF and VF, designed to take advantage of the unique characteristics and pollutant removal capabilities of different CWs types, by combining them in a strategic manner. These hybrid systems are often used to achieve specific pollutant removal goals more effectively. Many combinations are possible, including subsurface HF followed by VF, VF followed by HF and other stages of filters including water recirculation from one stage to another. This combination leverages the advantages of both HF and VF systems to achieve comprehensive treatment. Hybrid CWs systems offer a versatile and efficient means of treating a wide range of pollutants found in WW. By combining the strengths of different CWs types and optimising the treatment process, these systems can be tailored to meet specific pollutant removal requirements and site conditions (Wallace et al., 2020).

CWs became a widely accepted technology to deal with both point and non-point sources of polluted waters as they offer a technical, low-energy, and low-operational-requirements alternative to conventional treatment systems, while if properly design can meet the most stringent discharge standards. Used initially to treat municipal WW, the application of CWs has been expanded to the treatment of industrial effluent, agricultural WW, livestock farm effluent, landfill leachate and stormwater runoff, among others. Industrial WW differs substantially in composition from municipal sewage, as well as among themselves (Torrens, 2015). Industrial WW can present very high concentrations of organics, total suspended solids (TSS), ammonia and other pollutants; therefore, the use of CWs almost always requires some kind of pre-treatment (Vymazal 2010).

1.3 Intensified systems

The HF and VF CWs are passive treatment systems that can be operated without external energy supply (with exception of pumps that might be required for loading). Over the last decade, new wetland designs and/or operational strategies have been developed in order to comply with higher water quality standards for phosphorus and nitrogen removal and to reduce surface area requirements. These new strategies have led to a group of wetland technologies that are collectively referred to as intensified treatment wetlands.

It has been demonstrated and well recognised that CWs can efficiently remove organics, considerably remove nutrients, heavy metals, pathogens, pharmaceutical components and contaminants of emerging concern, including antibiotics and antibiotic resistance genes (ARGs) through enhanced configuration and operational strategies. Despite significant advancements, the widespread application of these systems is still limited by their current footprint and operating efficiency. Therefore, it is crucial to gain a comprehensive understanding of operation and design of CWs to improve the implementation of these technologies (Zhang et al., 2018).

Important factors affecting the treatment performance include the flow type, substrate characteristics, plant species, hydraulic loading rate (HLR), surface loading rate (SLR) and temperature. HLR and SLR, are the principal parameters for the design and operation of CWs. Temperature affects the metabolic activity of microorganisms in CWs. Warmer temperatures generally increase the rate of biological processes, such as the decomposition of organic matter (Wallace 2014).

The aerobic processes that require high oxygen availability can be limited in CWs. For Horizontal subsurface flow CWs, if nitrification (and subsequent total nitrogen removal) is a treatment objective oxygen might be a limitation due to the nature of the system. While the presence of macrophytes in CWs can enhance oxygen availability through the diffusion of oxygen via aerenchyma to the rhizomes, the contribution of plants to oxygen levels is generally deemed low, particularly in CWs receiving high SLRs. (Wießner, et al., 2002). As a result, to increase the oxygen availability, an option is to install an artificial aeration system. In this way, CWs design variants range from completely passive systems (HF, and also VF systems) to moderately engineered systems (unsaturated VF systems with pulse loading) up to highly engineered or intensified systems (Table 2). Numerous studies have found that adequate oxygen levels can enhance the treatment wetland efficiency and reduce its footprint (Fu, et al., 2023, Zhang et al., 2014, Zapater-Pereyra et al., 2014, Zhang et al., 2010, Wallace and Nivala 2008). It was in this context that aerated CWs have been developed. Aeration strategies have been introduced in CWs design to address limitations in oxygen availability (for example, limited nitrification in HF CWs).

Although the knowledge published in international journals and scientific literature on the enhanced treatment performance of intensified CWs has increased in recent years, aerated CWs design criteria continue to be largely a matter of patents and commercial practices.

Table 2: Intensified strategies.

Type of intensified input	Intensification Class	Example
Energetic	Aeration	Aerated Treatment wetlands
	Pumping	Reciprocating Fill and Drain TWs
Physicochemical	Sorptive media	Slags, expanded clay, zeolites, bauxsol, chitinous material, engineered materials
	Chemical dosing	Alum, ferric chloride, oxidising agents
Operational	Frequent plant harvesting	Duckweed systems, willow systems
	Cyclical resting	Routine alternation between multiple TW units in parallel
	Recirculation of flow	Vertical flow TW with recirculation
Microbial electrochemestry	Electroactive bacteria	METlands

1.3.1. Aerated Constructed Wetlands

The introduction of oxygen in CWs can enhance their treatment efficiency, especially for processes such as nitrification and denitrification. There are several mechanisms to introduce oxygen into CWs. The natural process of photosynthesis by aquatic plants generates oxygen during daylight hours. The roots of the plants also provide oxygen to the rhizosphere, enhancing microbial processes. Incorporating aquatic plants like emergent or floating vegetation can introduce oxygen into the system. Artificial aeration involves the use of mechanical devices, such as diffusers or surface aerators, to bubble air or pure oxygen into the water within the wetland (Freeman et al., 2018).

Aerated CWs are intensified systems in which air is injected at the bottom of a water saturated, media filled, basin. The aeration system consists of an air pump connected to a subsurface network of air distribution pipes to introduce air bubbles into the water (Wallace 2001). The use of an artificial aeration system increases the oxygen transfer rate, when compared to passively aerated CWs (e.g. VF) enabling improved performance for treatment reactions that require oxygen (such as organic matter removal and nitrification) that will occur faster under aerobic conditions. Additionally, the aeration system can also be operated intermittently to enhance total nitrogen removal through nitrification/denitrification (Figure 2).

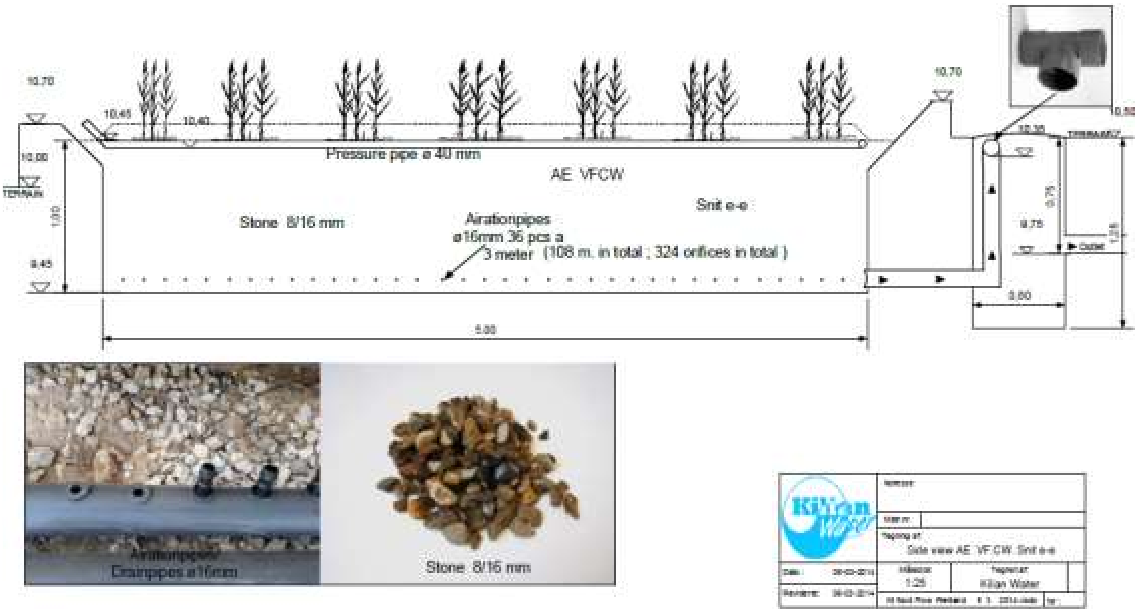


Figure 2: Lateral view of the vertical flow bed fitted with aeration system (Arias et al., 2015).

According to Dotro et al., 2017 and Vymazal 2017, aerated CWs have higher removal efficiency for nitrogen and carbon as well as pathogens, compared to non-aerated wetlands.

Furthermore, aerated CWs are flexible if variation of available oxygen is desired (e.g total nitrogen (TN), naproxen, diclofenac removal). The aeration strategies can vary from partial to total aeration in relation to time and space, and from low to high intensity depending on the contaminants targeted. The augmentation in dissolved oxygen by the application of artificial aeration improved the removal of pharmaceuticals, which are degraded under aerobic conditions. (Ilyas H, and van Hullebusch (2020).

However, this strategy requires automated devices and, usually, continuous aeration was provided, over the overall wetland bed or only near the inlet zone. However, a timetable for intermittent aeration or spatial segregation of aerated and non-aerated zones has been considered convenient in order to reach simultaneous nitrification and denitrification. In an efficient aerated HF CW, nitrification occurs when the aeration system is turned on, while denitrification requires anoxic conditions which could be obtained by ceasing aeration. Denitrification also requires a carbon source which must be furnished by the influent WW substrate. Thus, effluent recirculation could be necessary to improve the contact between the generated nitrate, the influent organic substrate and microbial population during anoxic periods (John et al., 2020).

Several parameters can affect the wetland aeration efficiency and, consequently, the performance of intensified treatment wetlands: gravel size and shape, plants presence, water temperature, bed and water depth, HLR, daily flow, surface organic loading rate, hydraulic retention time (HRT), airflow rate, total aeration time per day, atmospheric pressure, WW quality, fouling phenomena, maintenance of diffusers and others. The bed depth is an important factor because it determines, among other aspects, the power demand of the air blower and the residence time of the air bubbles in the system and consequently the O_2 transfer rates.

As pointed out by Rous et al. (2019), up to now the insufficient information on the aeration systems is quite common in research articles. From 21 reviewed of systems reported in the literature with an average bed depth of 0.53 m, Rous et al. (2019) found no correlation between water depth and aeration efficiency or removal efficiency. The effect of bed depth was one of the research topics of this thesis.

1.3.2. Recirculation

Recirculation involves returning treated water and mixing a portion of the wetland effluent with the influent of the treatment plant. Effluent recirculation has been proposed as an operational modification to improve organic matter and nitrogen removal especially in highly aerobic VF systems (Brix & Arias 2005). Removal of TN is enhanced because effluent with nitrate, but limited organic matter is mixed with influent low in nitrate but high in organic carbon and anoxic conditions, allowing denitrification to take place (Torrijo et al., 2016).

Though most common with VF systems, recirculation has been applied to virtually all treatment wetland types. As early as the 1990's, experiments with recirculation on a VF wetland showed improved efficiency and resulted in recirculation being incorporated into some of the design guidelines (Laber et al., 1997; Brix and Johansen, 2004; Brix and Arias, 2005). Recirculation ratios range generally from 50% to 200% of the influent flow. Higher ratios return more nitrate for additional denitrification but simultaneously increase the hydraulic loading and therefore decrease the HRT of the first pass influent, thus the proper recirculation ratio is specific to the hydraulic and nutrient loading rates of the system (Dotro et al., 2017).

Effluent recirculation (ER) is an effective strategy, which allowed adequate contact between the WW and biofilm. However, it remains unclear under which operational conditions the benefits of ER are optimal and if sufficient to justify the additional costs.

1.4 Types of WW

HF wetlands are used for secondary and tertiary treatment of domestic WW, as well as for a variety of industrial effluents such us petrochemical, chemical industry, paper and pulp WWs, abattoir, textile industry, tannery industry, food industry, distillery and winery, pig farm, dairy, airport runoff, etc (Vymazal 2010; Kadlec and Wallace, 2009). For HF wetlands treating domestic WW, primary treatment is generally achieved via a septic tank or an Imhoff tank. These systems are widely used in the Czech Republic, Spain, Portugal, Nicaragua, and North America among other countries for secondary treatment of domestic WW (Vymazal and Kröpfelová, 2008). In warm climate regions, it is common to find HF wetlands following septic tanks, anaerobic baffled reactors (ABR) and UASB reactors. In the UK, HF wetlands are predominantly used for tertiary treatment, with over 600 HF wetlands in operation. In this scenario, secondary treatment is often achieved using biological treatment units such as rotating biological contactors or trickling filters, and the HF wetlands are used as a polishing step. Additionally, combinations of HF with other wetland types (VF, FWS) have been used in a variety of hybrid systems (Dotro et al., 2017).

VF wetlands gained prominence in the 1990s in response to the change of the discharge requirements in Europe (specifically in Austria, Denmark, and Germany) that required elimination of ammonia nitrogen for small WW treatment plants. A large number of variants of VF wetlands exist (Stefanakis et al., 2019), including French VF wetlands treating raw WW and sludge TWs. VF wetlands are commonly used for secondary as well as tertiary treatment of domestic WW. VF wetlands are also used to treat landfill leachate and food processing WWs, which often contain high levels of ammonium nitrogen and/or organic carbon (upwards of hundreds of milligrams per litre) (Kadlec and Wallace, 2009), as well as other agro-industrial WWs such as olive mill effluents, dairy farm WW and animal farm effluent (Stefanakis et al., 2019).

In summary, CWs technology is versatile and environmentally friendly, and suitable for treating different types of WW. However, it has limitations in terms of the large area requirement and gravel bed clogging.

The use of CWs to treat industrial WW has increased over the past ten years (Rossmann et al., 2013). Industrial WW composition differs considerably from that of municipal sewage and is quite variable in itself. Unlike municipal WW effluents, which usually have a similar composition, industrial WW tends to have a variety of components with varying degrees of biodegradability and toxicity, thus requiring different treatment designs and strategies. In many industrial WWs the concentrations of organics, suspended solids, ammonium or other pollutants are quite high and therefore, the use of TWs nearly requires some sort of pre-treatment. In addition, there are no general rules for selecting the most suitable type of CWs for industrial WW treatment or even for urban WW (Torres A. 2015).

To overcome this limitation and enhance treatment efficiency, forced aeration in aerated CWs systems is used. The specific design of CWs systems should be based on the characteristics of the WW and the regulatory standards in the area, and it may require innovative approaches to address the challenges posed by varying conditions and pollutant loads.

Studies on industrial WW treatment on artificially aerated CWs are scarce. Results for coffee processing WW (Rossman et al., 2015), dairy parlor WW (Tunçsiper et al., 2015), aquaculture effluent (Zhang et al., 2015) and dye containing WW (Ong et al., 2011) are summarised below.

Rossmann et al., (2013) treated coffee processing WW (CPW) with aerated and non-aerated influent previously to pilot-scale HF CWs. The applied organic load during the experiment was 89 g COD/m^2/d, and the HRT was 12 d. Removal efficiencies of COD, BOD_5 and TSS ranged from 87.9 to 91.5, from 84.4 to 87.7 and from 73.7 to 84.8%, respectively. Aeration of CPW in the storage tank for 2.5 days did not affect the removal efficiencies of organic matter in the CWs, which agrees with previous findings of Zhang et al. (2010), due to low redox values and anoxic conditions in spite of the aeration. In this study, phosphorus removal (54.3–72.1%) was statistically different among treatments, with better performance for the aerated planted system, and worse for the non-aerated unplanted.

The feedlot runoff and dairy parlor WW in Burlington (Vermont, USA) was treated in four HFCWs (non-aerated unplanted CW1, aerated planted CW2, non-aerated planted CW3, and aerated planted CW4) of 225 m^2 each in an experiment carried out by Tunçsiper et al (2015). HRT in CWs ranged from 3 to 16 days. Over the four years of monitoring, the CWs operated with SLR of 210 g $BOD_5/m^2/d$ and 70 g $TSS/m^2/d$ in average. Average BOD_5 removals were 83%, 78%, 84% and 86% for CW1, CW2, CW3 and CW4, respectively. The introduction of supplemental aeration had a positive impact on BOD5 reduction. Aerated CWs exhibited higher BOD_5 and TSS treatment efficiencies by 8% and 5%, respectively, compared to non-aerated ones (Tunçsiper et al (2015).

The performance of the CWs showed improvement with increasing operational age and during optimal plant growth seasons, typically occurring between April and September. The adoption of an in-series design further increased BOD_5 and TSS removal efficiencies by 12% and 16%, respectively.

Aquaculture effluent under high HLR was assessed in Jingzhou city (China) by Zhang et al (2015). Two parallel, identical hybrid wetland systems (CW 1+2), each with down, up and HF chambers were constructed in the field. The HLR was approximately 8.0 m/d, giving a theoretical HRT of 0.96 h. For the wetland with diffused-air enhancement, there was a significant decrease in COD and NH_4^+-N concentrations. Further, the aeration significantly increased the levels of DO. This thesis discusses the use of CWs in various configurations for treating different types of WW, including urban and industrial WW from food and winery facilities.

1.5 Pollutant and pathogen removal processes

CWs are complex WW treatment systems with diverse pollutants and pathogen removal pathways. Macrophytes plays a very important role in removing various types of contaminants. Contaminants uptake by root zone involves rhizodegradation process by microbial activity. The heavy metals are reduced through phytostablisation. Plant enzymes break down the contaminants through phytodegradation. Plants and algae remove the contaminants from soil and sediments through phytoextraction. Plants release the contaminates from soil and sediments in atmosphere through phytovolatalisation process.

1.5.1 Organic matter removal

Removal mechanisms for particulate and soluble organic matter differ and depend on treatment wetland design. Generally, COD is used as the main analytical method for measuring organic matter, however, 5-day (carbonaceous) Biochemical Oxygen Demand (BOD_5) can also be used. BOD_5 is removed by biological degradation, sedimentation, and microbial uptake – organic contamination including pesticides are removed by adsorption, volatilisation, photolysis, and biotic degradation, suspended solids are removed by sedimentation and filtration. Nitrogen can have several pathways, including plant uptake, sedimentation, nitrification/ denitrification, microbial uptake, volatilization, Phosphorus is removed by sedimentation, filtration, adsorption, plant and microbial uptake, pathogens are removed by natural die-off, sedimentation, filtration, UV degradation, adsorption, heavy metals are removed by sedimentation, adsorption by vegetation and substrate and plant uptake process (Sánchez et al., 2022, Swarnakar et al., 2022).

Some WW parameters such as COD and BOD_5 provide bulk measurements of a range of organic compounds of varying degradability. Some compounds are more easily (or more quickly) degraded, and others are more difficult (or slower) to degrade. Therefore, the organic matter in the influent WW has a different composition than the organic matter that remains in the effluent (Wallace and Knight, 2006).

Two additional points that must be addressed since there are not complete solved when working with CW construction is clogging prevention and phosphorus removal.

1.5.2 Nitrogen removal

Nitrogen compounds in WW is one of the principal constituents of concern due to their role in eutrophication and effect on oxygen concentration in receiving waters (Kadlec and Wallace, 2009). Nitrogen exists in various forms including organic matter, NH_4, nitrate (NO3), nitrite (NO2), or nitrogen gas, depending on the oxidation/reduction conditions of the CW (Wallace & Knight, 2006). Removal mechanisms of N from CWs include ammonia volatilization, denitrification, uptake by vegetation followed by biomass harvesting, and ammonia adsorption (Vymazal, 2007). Other processes occurring in CWs such as nitrification [a process mediated by microbes which is an important mechanism to reduce the concentration of ammonia], are responsible for converting N to various forms, but do not remove N from WWs (Vymazal, 2007). However, nitrification coupled with denitrification (a temperature dependant process which is also dependant on the availability of organic C, in which the oxidised N compounds, nitrate (NO_3^-) or nitrite (NO_2^-), are reduced to the N gases - N_2 or nitrous oxide (N_2O)), appears to be a major N removal mechanism in CWs (Vymazal, 2007).

1.5.3 Phosphorus removal

Phosphorus enters most treatment wetlands primarily as organic phosphorus and orthophosphate, but most organic phosphorus is converted to orthophosphate as part of organic matter degradation. Mechanisms that play a part in phosphorus removal in treatment wetlands include chemical precipitation, sedimentation, sorption and plant and microbial uptake. Unfortunately, most of these processes are slow or not active unless special media are used to enhance abiotic processes.

The effectiveness of treatment wetlands for phosphorous removal is determined by the applied loading rate. In very lightly loaded FWS systems, such as for effluent polishing, phosphorus removal can be excellent and due primarily to soil accretion (sedimentation and co-precipitation with other minerals). For treatment of typical secondary WW using VF and HF systems, removal is generally quite modest once the adsorptive capacity of the media is saturated (Dotro et al., 2017).

Among these processes, adsorption and precipitation seem to play the largest role in phosphate removal in CWs. To ensure efficient phosphorus removal, research should aim to identify substrates that have a high phosphorus removal capacity, and suitable properties for use as CW substrate. Particularly, there is a great interest in studying the beneficial reuse options of resource recovery which can be an alternative to gravel, which can contribute to the improvement of WW treatment systems.

1.6 Critical factor: clogging

The most critical operational issue for HF wetlands is clogging. This occurs when the pore spaces in the media are filled with solids (organic or inorganic, thus limiting the contact area and time between the biofilm and the water. Clogging can occur in any kind of (biological) filter and has been reported for both HF and VF systems (Knowles et al., 2011). For HF wetlands providing treatment of domestic WW, clogging is most commonly caused by excessive organic and/or solids loading onto the gravel bed. This is often due to improper maintenance of the septic tank (secondary treatment HF wetland) or final settling tanks (tertiary HF wetlands), and/or poor dimensioning of the wetland itself. Hydraulic and solids loading rates that are at the top end of recommended values have been suggested as the main factors resulting in the reported clogging of HF systems.

Two additional points that must be addressed since there are not complete solved when working with CW construction is clogging prevention and phosphorus removal. One of the main problems with operating subsurface HF systems is clogging the media. Although the clogging phenomenon is an extremely complex and not well-understood process, the influent content in suspended solids is an important factor. To minimise the risk, the use of anaerobic digesters as pre-treatment can achieve high suspended solids removal and contribute to prevent clogging problems. Additionally, the combination anaerobic digesters followed by a CW is sound since both technologies agree with the criteria of low cost and sustainability, including simplicity of construction, operation and maintenance (de la Varga et al., 2013a).

2. List of Papers

This doctorate thesis is comprised of the following papers, which are referred in the text by their Roman numerals (Table 3) .

Table 3: List of papers in this thesis.

Roman numerals	Title
I	Pascual A., De la Varga D., Arias C.A., Van Oirschot D., Killian R., Álvarez, J.A., Soto, M., 2016. *Hydrolitic anaerobic reactor and aerated constructed wetland systems for municipal WW treatment – HIGHWET Projec*t. Environmental Technoloy. https://doi.org/10.1080/09593330.2016.1188995
II	Pascual A., De la Varga D., Soto M., Van Oirschot D., Kilian R.M, Álvarez J.A., Carvalho P. and Arias C.A., 2018. *Aerated Constructed Wetlands for Treatment of Municipal and Food IndustryWW. ConstructedWetlands for IndustrialWW Treatment,* First Edition. Edited by Alexandros Stefanakis. © 2018 JohnWiley & Sons Ltd. Published 2018 by JohnWiley & Sons Ltd. DOI:10.1002/9781119268376.ch3
III	Pascual A., Álvarez J.A., De la Varga D., Arias C.A., Van Oirschot D., Killian R., Soto, M.*, 2023.Horizontal flow aerated constructed wetlands for municipal WW treatment: the influence of bed depth.* Science of the Total Environment. https://doi.org/10.1016/j.scitotenv.2023.168257
IV	Pascual A., Pena R., Gómez – Cuervo S., De la Varga D., Álvarez J.A., Soto M.*,* Arias C.A 2021. *Nature based solutions for winery WW valorisation.* Ecological Engineering. DOI: 10.1016/j.ecoleng.2021.106311

3. Objectives

The **overall objective** of this thesis is to investigate and optimise the factors affecting WW treatment in aerated CWs receiving various types of WW, including urban and industrial WWs, with the goal of improving the design parameters and future implementations of aerated CWs for WW treatment. Additionally, the thesis aims to assess the feasibility of the HUSB reactor followed by CWs configuration for treating WW from diverse sources, such as urban, food industry, and winery WWs, and to evaluate the impact of design and operational parameters on treatment efficiency.

Research findings were aimed to increase knowledge about proper guidelines for CWs design, operation and maintenance. A combination of HUSB reactor and CWs to treat urban, high load organic industrial and winery WW were studied during several years. Selected critical factors were units performance, phosphorus removal, HUSB reactor for pre-treatment to prevent clogging risks in CWs, the influence of bed depth in aerated CWs, treatments efficiency.

This research is relevant, impact, importance to improve and the design of efficient and economic aerated CWs systems. There is still a need for an improved understanding of the internal processes involved in this type of systems. Further, performance data and information that can guide design and operation of aerated CWs is scarce. These issues are addressed in the thesis.

The specific objectives of these thesis are showed in Table 4.

Table 4: Specific Objectives and papers that are included in the present doctorate thesis.

Paper number	Specific Objective	Geographic location
I, II, IV	To evaluate performance efficiency of the HUSB - aerated CWs configuration on urban and industrial (food and winery) WW treatment efficiency.	Spain, Denmark
I, II, IV	To validate the configuration HUSB-aerated CWs and quantify their function for pollutants removal potential with emphasis on nutrients (TN) and total phosphorus (TP)).	Spain, Denmark
I, II	To obtain optimal aeration regime to treat urban and food industrial WW.	Spain, Denmark
IV	To examine the influence of design parameters and operational parameters (organic and SLR) on the treatment efficiency of HUSB –CWs configuration.	Spain
I	To reduce the required area for the CWs.	Spain
I, II, III, IV	To prevent CW clogging using a HUSB as pre-treatment.	Spain, Denmark
I, II	To study different phosphorus adsorbent materials for the treatment of urban and food industrial WW.	Spain, Denmark
III	To study the role and influence of bed depth in aerated CWs to treat urban WW.	Spain
I, IV	To validate the combination of HUSB and CWs to treat food industrial and winery WW.	Spain, Denmark

4. Methods

4.1 Study sites and experimental designs

The studies presented in this thesis have been carried out in Spain and Denmark in two pilot plants and one full scale plant:

- **HIGHWET pilot plant** located at the outdoors of the Science Faculty of the University of A Coruña (A Coruña, Spain). This pilot plant was operated and study for 2 years and the results are presented in papers I and III (Figure 3 and Figure 4).

Figure 3: Geographical location of the HIGHWET pilot plant at the UDC facilities in A Coruña (Spain).

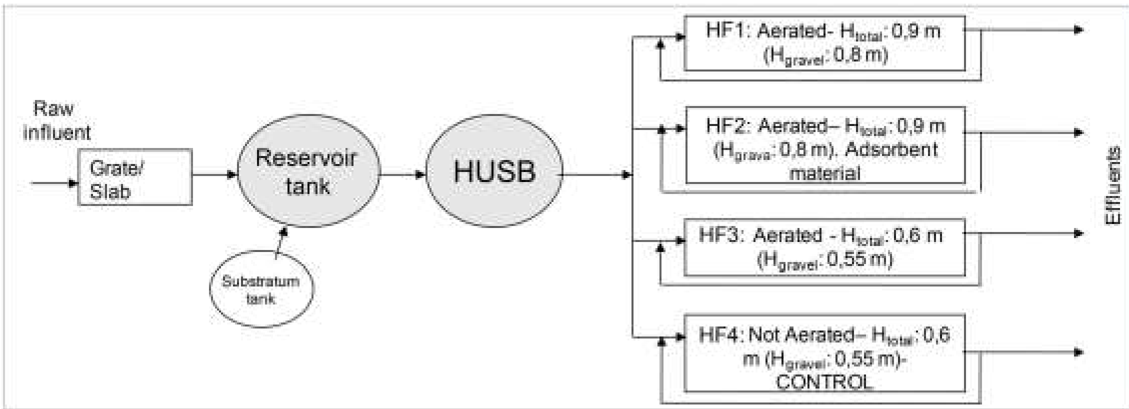


Figure 4: Flowchart and CWs of HIGHWET pilot plant.

- **KT Food pilot plant** located nearby Aarhus (Denmark). The pilot plant was operated and studied during Paper IV (Figure 5).

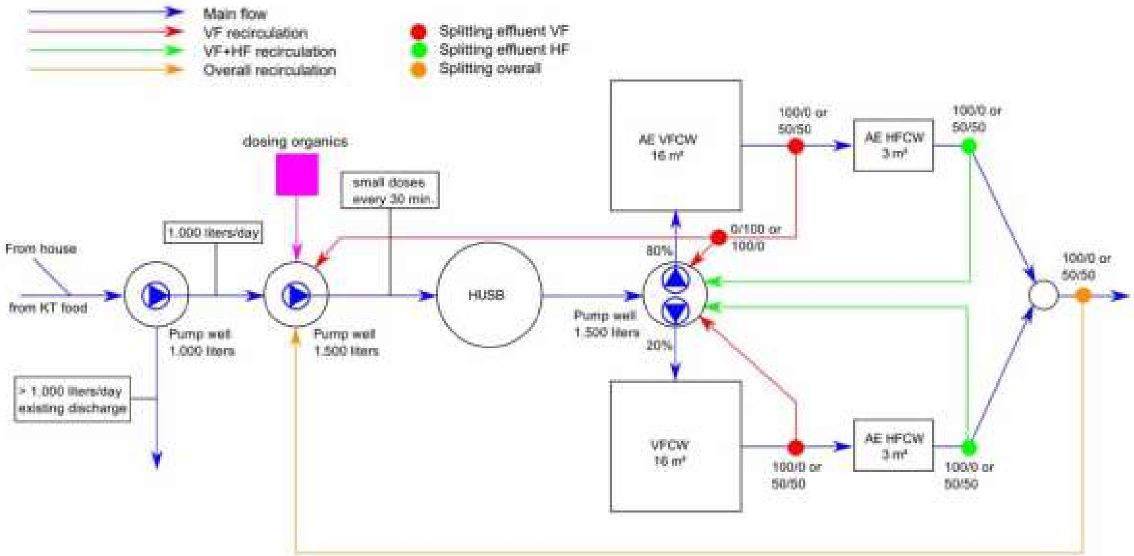


Figure 5: Geographical location of the factory, the house and a flowchart of pilot plant at Aarhus (Denmark).

- **WETWINE plant** located in the facilities of the Santiago Ruiz Winery in Tomiño (Pontevedra, Spain). This pilot plant was operated and study during the paper IV (Figure 6 and
- Figure 7).

Figure 6: Geographical location of the WETWINE pilot plant at Tomiño (Spain).

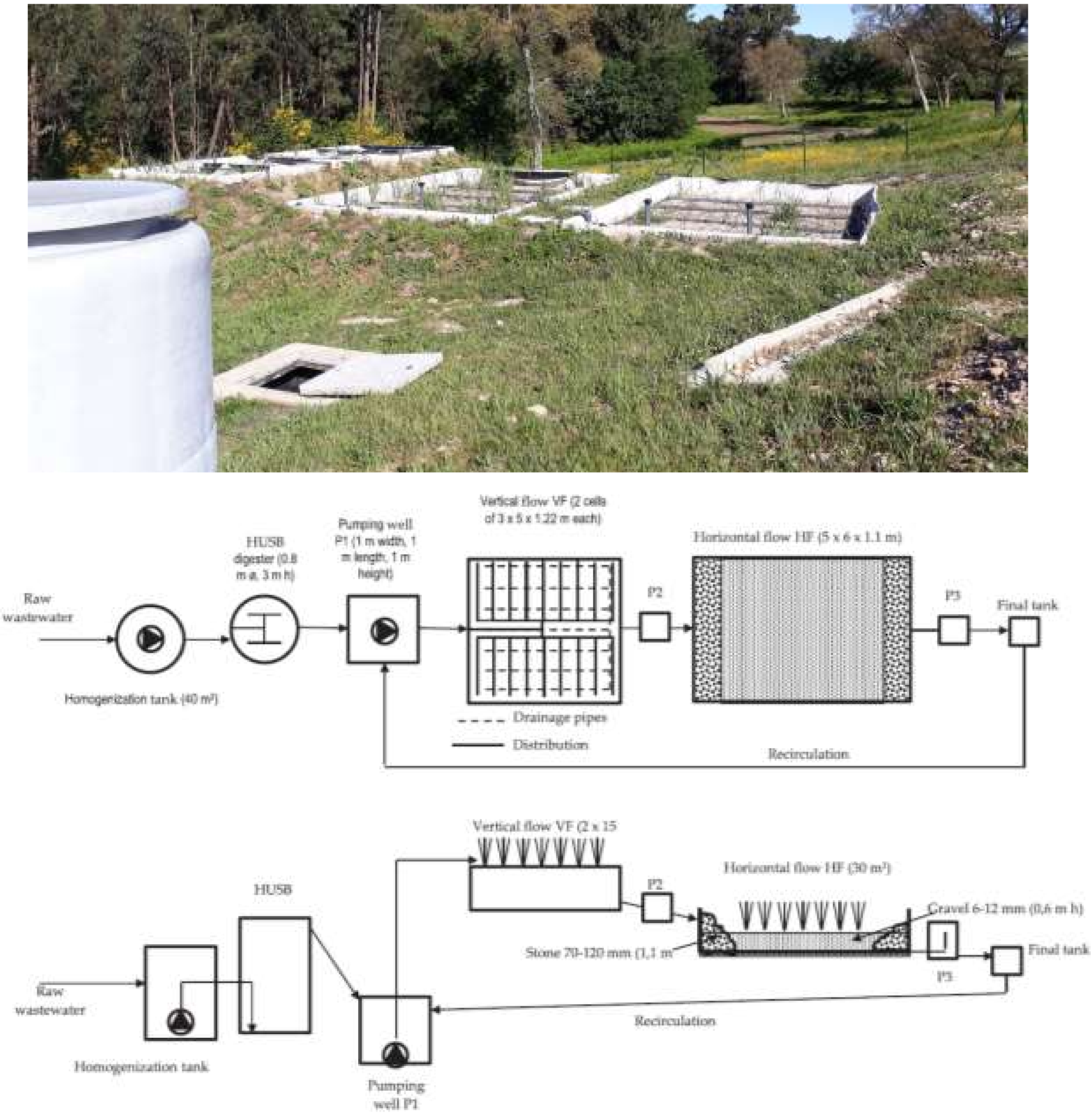


Figure 7: Photo and flowchart of WETWINE pilot plant.

The research published in articles I, II and III was carried out in two pilot plants: the HIGHWET pilot plant (Spain) and the KT Food pilot plant (Denmark).

The first configuration (HIGHWET pilot plant) located in A Coruña, Spain (43° 19' 36.444" N 8° 24' 31.068" W) consisted of a HUSB reactor followed by four HF CWs working in parallel and receiving anaerobic pre-treated WW. HF1, HF2 and HF3 units are fitted with aeration, while the HF4 is not aerated in order to be used as a control (paper I). All the HF units were provided with effluent recirculation, but different heights of gravel bed were implemented in order to compare the behaviour of the aeration operating at high organic and hydraulic load in different scenario for raw municipal WW treatment (paper III).

The second configuration (nearby Aarhus, Denmark) consisted of a combination of a HUSB reactor as primary treatment, followed by two parallel treatment trains (aerated line and non-aerated line) of hybrid CWs (VF and HF), several wells to allow controlled recirculation of treated waters and additional wells to host reactive media to remove P before discharge (paper II). This configuration was designed for the treatment of high-load organic industrial WW. KT Food is a food producing company located at Randersvej in the town of Purhus (56º 33'38.58" N 9º 51' 26.08" E). Additional to the production of food, the site also generates water from a dairy farm and domestic activities.

The study of Paper III was performed in an experimental pilot plant in the facilities of the Santiago Ruiz Winery in Tomiño, Spain (42°34'41.50" N • 8°44'01.50" O). The winery has 35 ha of planted vineyard; it produces 368.000 L/y of wine resulting in large volumes of highly loaded WW. During the two months of the grapes harvest, the estimate water produced is 620 m^3 while during the one-month grape fermentation period the WW produced reaches 130 m^3 for the remaining 9 months of off-peak period, the volume of WW and the pollutant load decreases considerably to a total of 648 m^3. The WW treatment line of the WETWINE pilot plant consisted of a HUSB reactor followed by two parallel unsaturated VF, and HF CWs (i.e. hybrid CWs).

5. Main results and discussion

HUSB reactor performance

Three additional points of interest about the performance of HIGHWET plants which are currently being assessed are those related to substrate clogging, greenhouse gases (GHG) and energy consumption. In paper I, the plant included an anaerobic pre-treatment step in order to reduce the entrance of suspended solids to HF beds and help in clogging prevention. Additionally, compared to conventional HF units, larger gravel was used in aerated units. Clogging can also be caused by biofilm development (Zhao et al., 2009) which is a risk in more intensive systems.

Preventing wetland clogging is crucial to maintaining the effectiveness of a CW for WW treatment or ecological purposes. Clogging can occur due to the accumulation of organic and inorganic matter, which reduces the flow and treatment capacity of the wetland. A pre-treatment is a strategy to help prevent wetland clogging. Implement pre-treatment processes to remove large debris, grit, and solids before the WW enters the wetland. This reduces the organic and inorganic load on the wetland.

An anaerobic reactor with a similar design to the typical up-flow anaerobic sludge blanket (UASB) reactor when it is used at hydrolytic (non-methanogenic) conditions is known as a HUSB reactor (Álvarez et al., 2008). The HUSB reactor should minimize methanogenic activity while enhancing hydrolysis and acidification of particulate matter in order to reach high volatile fatty acid concentrations in the effluent. Therefore, a HUSB reactor is mainly used for that WW with a high suspended solid concentration in order to solubilise particulate matter and to increase the removal of easily biodegradable organic matter in the WW. Treating municipal WW, the HUSB reactor can operate at HRT ranging from 3 to 14 h (Álvarez et al., 2008).

In the HIGHWET pilot plant (paper I), the HUSB was operated at approximately 6 h of HRT. The WW parameter most affected by the anaerobic treatment is the particulate organic matter. The HUSB removed 76–89% of TSS, highly reducing the influent concentration of suspended solids to the HF units. Average Chemical Oxygen Demand (COD) and BOD_5 removal was 42% and 48%, respectively. Additionally, the HUSB digester removed organic nitrogen (Org. N, calculated as the difference between TN and ammonia and nitric nitrogen) at a rate ranging from 14% to 22% and increased the ammonia concentration by about 35%. Available data from periodic inspections of the height and concentration of the sludge bed indicate a rate of sludge formation of approximately 2 L/d with a concentration of about 30 g VSS/L and 60 g COD/L. A COD balance indicates that the sludge bed accumulates about 6% of influent COD, which in turn is about 14% of the removed COD. Once the maximum sludge bed of about 1.2 m is reached, maintaining the steady state will require the purge of about half (or more) of the accumulated sludge at each three months period.

The results obtained in KT Food pilot plant regarding TSS removal were lower. The paper II shows that the TSS reduction in the HUSB was around 60%. Nevertheless, a high reduction of COD occurred in the HUSB where 50% of the COD was removed. Denitrification occurred in the HUSB where 43% of the NO3-N was removed.

In the case of WETWINE pilot plant (paper IV), the HUSB effectively removed the TSS during several periods, with over 50% removal in all cases. During other periods, the TSS removal was low or even negative suggesting release of TSS. The low or negative TSS removal can be attributed to a high up-flow velocity in the HUSB (up to 4.4 m/h).

The results of the research reported in paper IV are in accordance with the results presented by De la Varga et al. (2013a) in a previous work on the application of HUSB for the treatment of wastewater from the wine industry. In their study, De la Varga et al. (2013a), stated that the HRT in the HUSB reactor ranged between 19 and 28 h, while in the WETWINE plant, the HUSB mostly operated at a HRT of 28 ± 13 h. Regarding volumetric loading rate of TSS (VLR_{TSS}), in both studies they ranged from 0.3 to 1.1 kg TSS/m3·d for high loading periods. For COD for the WETWINE plant VLR_{COD} the loading to the digester averaged 3.2 ± 4.4 kg COD/m3·d, higher than the one reported by De la Varga et al. (2013a) of 2.4 ± 1.6 kg COD/m^3·d. The volumetric loading rate of COD VLR_{COD} was particularly high during part of the study, reaching 14Kg COD/m^3·d. In the study by De la Varga et al. (2013a), the HUSB removed on average 76% TSS and 26% COD, somewhat better results than those found for the WETWINE project (paper IV), but that might be justified by the lower TSS and higher COD concentrations in the influent to the WETWINE plant.

The use of a HUSB reactor as an anaerobic pre-treatment step in the WW treatment process is effective in reducing the TSS concentration and in solubilize particulate matter. By reducing TSS in the influent WW, it is considered that the HUSB reactor helps prevent clogging in subsequent treatment units, such as CWs (Álvarez et al., 2008; De la Varga et al., 2013a). Clogging, often caused by the accumulation of organic and inorganic matter, can hinder the flow and treatment capacity of these units. Therefore, the role of the HUSB reactor in removing suspended solids is crucial for maintaining the effectiveness of the treatment process and preventing clogging issues.

In additions to the accumulation of influent TSS, clogging can also be caused by biofilm development (Zhao et al., 2009) which is a risk in more intensive systems. Thus, compared to conventional HF units, larger gravel was used in aerated HF units. The operation of the different units that received the pre-treated influent were monitored for long periods of time, between 1 and 2 years, without any clogging being observed (papers I-IV).

Organic matter removal in CWs

Papers I, II, IV provided a discusses the results and performance of the combination of HUSB and aerated CWs and non aerated CWs for the treatment of urban and industrial (food and winery) WW, with a particular focus on the impact of aeration on the removal of organic matter..

Treating domestic WW (paper I), the SLR ranged from 29 to 47 g $BOD_5/m^2 \cdot d$ (50–63 g $COD/m^2 \cdot d$) for the aerated units and from 8 to 14 g $BOD_5/m^2 \cdot d$ (14–19 g $COD/m^2 \cdot d$) for the non-aerated unit. The average surface removal rates (SRRs) were significantly higher in the aerated units, with SLRs of 37 g $BOD_5/m^2 \cdot d$, compared to 10 g $BOD_5/m^2 \cdot d$ in the non-aerated unit.

The results obtained during the research of paper I highlight the impressive performance of aerated CW systems in removing organic matter (TSS, BOD_5, and COD) from urban WW. The aerated lines in the study achieved exceptional removal of influent organic matter. The concentrations of TSS, BOD_5, and COD in the effluent were extremely low, often close to zero or below the detection limit. Removal percentages higher than 96% were consistently observed in the aerated units, indicating the high efficiency of these systems in treating organic matter. In comparison, the non-aerated HF unit had slightly lower performance, with removal rates of 90%

This suggests that aeration significantly enhanced the removal of organic matter compared to the non-aerated unit. Both the aerated and non-aerated units demonstrated sufficient capacity to remove organic matter effectively.

In the case of KT Food pilot plant (paper II), the SLR ($g/m^2/d$) in the aerated line (VF + VF) were 2.5 ± 1.8, 92 ± 14, 58 ± 7, 9.1 for TSS, COD, BOD_5 respectively, whilst SLRs were four times lower in the non-aerated line (VF + HF) (i.e., 0.6 ± 0.4, 23 ± 4, 15 ± 2 for TSS, COD, DBO_5). Thus, the non-aerated line operated at conservative design loading rates and reached satisfactory contaminant removal, usually from 90 to 99% of TSS, COD, BOD_5 and ammonia. Similar or even higher percentage removal rates were obtained in the aerated line, operated at four times higher loading rates.

The SLR generally recommended for secondary treatment in HF CWs is in the range of 4–6 g $BOD_5/m^2 \cdot d$. (Akratos and Tsihrintzis, 2007, Carballeira et al., 2016) So, the surface land required ranges from 5 to 7 m^2/pe. Compared to the non-aerated HF unit, aerated units in the paper I reached 3.7 and 5.5 times higher BOD_5 and TN SLRs, somewhat higher than those recently reported by Zapater-Pereyra et al., for aerated HF systems. Even the BOD_5 SLR in the aerated units was about eight times higher than the current design criteria for conventional HF units. Thus, we can conclude that the required area can be reduced by a factor of 5. These results could favour the extension of CW technology to serve WW discharges in a broad range above 2000 pe, namely in the range of populations from 2000 up to 5000 pe. In this range of application, septic tanks are not useful and HUSB digesters can clearly compete with Imhoff tanks and other WW pre-treatments.

The percentages obtained in WETWINE pilot plant (paper IV) for the combination of HUSB and CWs were also satisfactory. The COD removal was generally high in the VF that achieved average values ranging from 47% to 96% for the different operation periods. These removal percentages are high, even though the treated WW (winery WW) in this particular pilot plant has a higher load than the WW treated in the other two pilot plants.

The paper IV discusses the evaluation of variables that affect the removal efficiency of COD in a VF CW to treat winery WW. The study examined the impact of various variables on the efficiency of COD removal in the VF CW, which was the first step of the hybrid CW system. These variables included COD influent concentration (CODin), surface loading rate for COD (SLR_{COD}), HLR. It was observed that the % CODr increased with the SLR_{COD} up to values of 100-160 g COD/m^2·d and decreased for higher SLR_{COD} values. Multiple regression analysis was performed using all the data, and a model was developed to explain % CODr as a function of CODin and SLR_{COD}. This model had a good fit ($R_2 = 0.70$) and significance ($p < 0.01$) for the two variables, explaining 70% of the variation in COD removal in the VF.

The results obtained in paper IV, suggest that COD removal efficiency in the VF system is primarily influenced by CODin and SLR_{COD}, while TSS removal appears to be influenced by filtration mechanisms. Other factors like HLR and HRT had limited or no impact on the removal efficiencies. The findings can be valuable for optimizing the performance of VF treatment systems, especially in the context of WW treatment from wineries (Figure 8).

The HF unit, which was the last unit of the hybrid VF-HF CW system, showed low removal of COD in the first part of the study and medium to high COD removal in the second part. The efficiency of COD removal varied under different loading conditions. The percentage of COD removal decreased when the load exceeded 30 g COD/m^2·d and during low loading periods. Performance was highest (62–77% COD removal) during periods with a SLR in the range of 6 to 28 g COD/m^2·d. When high load periods were excluded from the analysis, a multiple regression model ($R^2 = 0.71$, $p < 0.05$) showed that COD removal increased with decreasing influent COD (CODin) and increasing SLR. HRT had a significant effect on COD removal ($R^2 = 0.75$, $p < 0.1$). As the HRT decreased, the effluent COD concentration increased.

Aeration significantly improved the removal of organic matter in the CW systems, leading to very low concentrations of TSS, BOD_5, and COD in the effluent

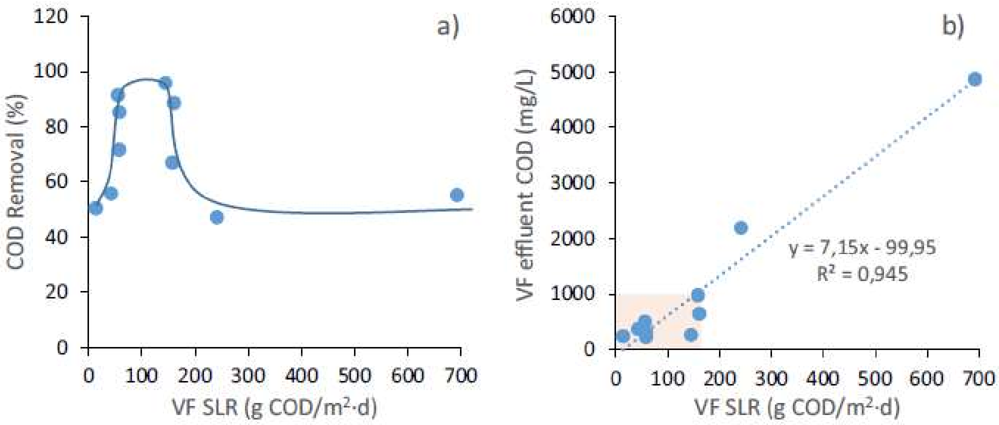


Figure 8: Effect of SLR on COD removal (a) and effluent COD concentration (b) in VF.

Paper IV highlights the importance of operational parameters, such as SLR and HRT, in WW treatment performance in HF CWs. The results obtained are on line with the study by Akratos and Tsihrintzis (2007). They studied the relationship between HRT and COD removal efficiency and heir results show that as the HRT decreases, the effluent COD concentration will increase. These results were confirmed by Trang et al. (2010), who observed the reduction in organic matter and nitrogen removal efficiency with the reduction of HRT in their system due to less contact time of contaminants in the wetland resulting in low effluent quality for reuse purposes in the agricultural sector.

Nitrogen

During this thesis the nitrogen removal was studied. In paper I the removal of several forms of nitrogen in the HUSB and HF units was calculated. The system removed organic nitrogen (Org. N, calculated as the disparity between total nitrogen (TN) and ammonia and nitric nitrogen) within a rate range of 14% to 22%, concurrently elevating the ammonia concentration by approximately 35%. Thus, the influent to the HF units (i.e. HUSB effluent) appeared mainly as ammonia nitrogen (71% and 85% of TN), the remaining corresponding to Org. N (15–28%), while nitric (NO_3^- and NO_2^-) nitrogen accounted for less than 1% of TN. All the aerated units effectively removed TN, Org. N and $NH4^+$-N at rates ranging from 47% to 100%, but differences existed mainly as a function of the unit configuration and the on/off aeration ratio. In addition, TN removal was improved by effluent recirculation in the aerated units, leading to 50–80% TN removal (Figure 9).

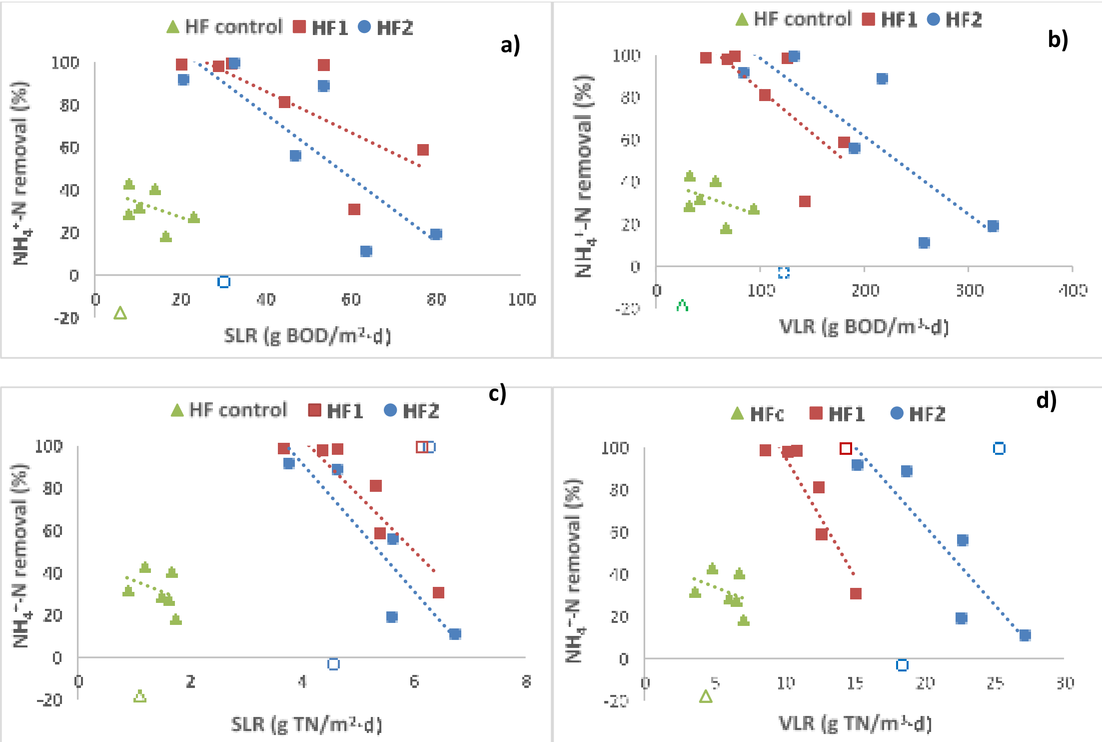


Figure 9: Correlation of NH4+-N removal with several loading rate parameters (non stuffed points are excluded from the equations).

Paper II analysed nitrogen removal in the HUSB and VF units at KT Food pilot plant. TN removal reached 43 % in the HUSB digester in the case of KT Food pilot plant due to the denitrification of influent nitrate. Overall, TN removal was 85 % in the non-aerated line and 91 % in the aerated line. The aerated VF CW unit reached 80 % TN removal whilst the non-aerated VF CW unit reached 58 % TN removal. The aerated line was successful in treating a four times higher loading rate and with similar or higher treatment efficiency than the non-aerated VF CW unit for nitrogen removal. These high percentage removal rates were obtained at TN loading rates higher in the aerated line of the KT Food HIGHWET plant (SLR ($g/m^2/d$) 9.1 ± 3.5, 7.8 ± 4.2 for TN andNH_4^+-N, respectively) in comparison of referred studies and to the non-aerated unit (SLR ($g/m^2/d$) 2.3 ± 0.9 and 2.0 ± 1.0 for TN andNH_4^+-N, respectively).

TN removal was dominated by ammonium removal in the aerated stage. In the non-aerated wetland, NH_4^+-N outlet concentrations were generally higher than in the inlet, observing an opposite trend in the aerated wetland. It can be concluded that denitrification process was contained with short HRT (0.96 h) even though carbon source seemed to be enough for denitrification. The results obtained in both pilot plants are in agreement with those obtained by other authors. The results obtained by Bodin, 2013 showed that the rise in dissolved oxygen possibly stimulated nitrification rates, contributing to the improved NH_4^+-N removal during this period.

The case of WETWINE pilot plant was different regarding nitrogen (paper IV). When compared to other similar reported study (Serrano et al., 2011), the concentration of nitrogen and phosphorus compounds in the WETWINE wastewater were low, with an average nitrogen concentration at the influent of 22.9 ± 23.5 mg TN/L, mainly in form of ammonium (15.9 ± 20.4 mg NH_4^+-N/L) and the rest as organic nitrogen. The VF unit reached 57% ammonia removal on average, while the HF effectively removed NO_2^- (87%) and NO_3^- (97%). TN removal reached 62% in the overall system, where at the final effluent the concentration was 7.0 ± 10.6 mg NH_4^+-N and 8.6 ± 11.9 mg TN/L.

Since nitrification and denitrification are two operationally separate processes (either temporally or spatially), which respectively require aerobic and anoxic conditions, the rate of nitrification significantly impacts the removal of TN. The removal efficiency of TN significantly dropped with an increase of HLR. Artificial aeration significantly improved the oxygen availability and thus enhanced the removal of NH_4^+-N in the VF CWs in the two pilot plants. Intermittent aeration was optimal for TN removal, which facilitated denitrification due to both spatial and temporal formations of anoxic zones in the VF CWs.

Different aeration regime for the aerated VF CWs were set to intermittent aeration in both pilot plants. HIGHWET pilot plant the aeration strategy were 5 hours on/3 hours off, 3 hours on/5 hours while KT Food pilot plant were 4 hours on/4 hours off and 6 hours on/2 hours off. The results from both papers indicated that aeration regimes had a significant influence on TN removal an in the WW treatment systems. This suggests that the aeration strategies played a crucial role in the overall performance and sustainability of the treatment processes. The findings highlight the importance of optimising aeration in CWs and related systems to achieve desired treatment outcomes. Aeration regimes ranging from 5 hours on/3 hours off to 3 hours on/5 hours off were found to be effective for optimising TN removal.

Otherwise, the results obtained during paper II showed that temperature can influence nitrification rates, denitrification, and plant nutrient uptake, all of which are critical processes for nitrogen removal in CWs. In colder months, the rates of nitrification and denitrification may decrease, potentially leading to reduced nitrogen removal efficiency. System operators and designers should consider seasonal temperature variations when designing and managing these systems to ensure consistent and effective nitrogen removal throughout the year.

Bed depth

Paper III highlights the impact of water depth, aeration, and specific loading rates on the performance of HF CWs for WW treatment. This paper shows a study that compared the performance of different HF CWs systems with varying water depths and aeration conditions for the treatment of urban WW.

The study compared an aerated CWs system with a water depth of 0.8 meters (HF1) to two other systems with a water depth of 0.5 meters, one of which was aerated (HF2), while the other served as a non-aerated control unit (HFc). The gravel size used in the systems was 12-16 mm, and the aeration rate was 0.69 $m^3/m^2 \cdot h$. The aeration rate used in this paper III was higher than that used by John et al. (2020) but lower than the range recommended by Vera-Puerto et al. (2022). Intermittent aeration was used in the study.

The control unit and the aerated HF1 unit demonstrated sufficient capacity to effectively cope with the applied SLR and produced high-quality effluent. The best COD removal, exceeding 90%, was achieved for HF1 when the SLR was up to 80 g $BOD_5/m^2 \cdot d$. In contrast, HF2 with a shallower bed depth and HFc achieved lower COD removal rates.

Aeration allowed for an increase in the SLR by a factor of 4 when the water depth increased from 0.5 meters to 0.8 meters. If the water depth remained the same, the SLR could only be increased by a factor of 2. This indicates that aeration has the potential to significantly reduce the surface area required for HF CWs, with percentage reductions ranging between 50% and 80%.

Phosphorus removal

Phosphorus removal in CWs is an essential aspect of WW treatment, especially in addressing nutrient pollution and eutrophication in natural water bodies.

The results obtained during the investigation discussed in paper I showed that The HUSB digester showed a minor effect on TP, but slightly increased phosphate content in the effluent.

In the non-aerated HFc unit, reduced removal of phosphate (about 20%) and total phosphorus (about 6%) was observed. These reduced removal rates are attributed to several factors, including the higher-than-recommended SLR applied (8-14 g $BOD_5/m^2 \cdot d$). In addition, monitoring campaigns took place during the winter (C1) and early spring (C2), during which the planted macrophytes were still dormant. The aerated HF1 and HF3 units also showed some reduction in phosphate and TP removal, although not as much as the non-aerated HFc unit.

In contrast to the other units, the tobermorite-enriched HF2 unit achieved notably high phosphate removal (60-67%) and TP removal (54%). This superior removal is attributed to the use of tobermorite material in this particular unit. Tobermorite is a type of mineral that has been shown to have the ability to adsorb and remove phosphorus from water effectively.

In the case of KT Food pilot plant, the SLR in the aerated line was 0.8 $g/m^2/d$ for TP. TP removal reached 39 ± 14% in the HUSB digester, whilst overall TP removal was 98 ± 1% in the non-aerated line and 90 ± 3% in the aerated line. Both the aerated and non-aerated VF CW units noticeable contributed to TP removal, reaching 72 ± 10% and 82 ± 13%, respectively. Additional TP removal took place in the aerated HF units, reaching 18 ± 12% and 42 ± 25% for HF1 and HF2, respectively. According to Vymazal (2007), these TP removal rates obtained under average loading rates of 0.8 g $TP/m^2/d$ may be considered very satisfactory. Finally, the P removal units with Polonite as phosphorus adsorbant material reached 56 ± 5% TP removal whilst TP removal in the unit with Tobermorite decreased from about 50% in the third month to 11% in the ninth month.

The effectiveness of phosphorus removal in CWs can vary depending on factors such as influent phosphorus concentrations, hydraulic loading rates, design characteristics, and maintenance practices. Proper design and management of CWs are essential to optimise phosphorus removal and ensure the long-term performance of these systems in treating wastewater and reducing nutrient pollution.

Hybrid CWs

The analysis of the paper II data showed that the combination of HUSB and hybrid aerated and non-aerated CWs was successful to treat food industrial WW. Applied SLR ($g/m^2/d$) in the aerated line were 2.5±1.8, 92±14, 58±7, 9.1±3.5, 7.8±4.2 and 0.8±0.1 for TSS, COD, DBO5, TN, NH_4^+-N and TP, respectively, whilst SLRs were four times lower in the non-aerated line (i.e. 0.6 ± 0.4, 23 ± 4, 15 ± 2, 2.3 ± 0.9 and 2.0 ± 1.0 for TSS, COD, DBO5, TN and NH_4^+-N, respectively). The non-aerated line reached satisfactory contaminant removal, usually from 90 to 99% of TSS, COD, BOD_5 and ammonia. Similar or even higher percentage removal rates were obtained in the aerated line. The results showed that the aerated VF CW was successful in treating a four times higher loading rate and with similar or higher treatment efficiency than the non-aerated VF CW.

The data analyzed for each unit during the operating time showed that the HUSB unit effectively removes about half of the TSS, COD and DBO. The VF CWs removed on average more than 99% of the remaining BOD_5 leaving very little BOD_5 to be removed in the following HF CWs. The HF CWs contributed to refine the removal of the remaining BOD_5 and COD. The nitrification process occurred mainly while the water was in the VF CWs. The VF unit reached 95% ammonia removal on average, while the HF effectively removed nitrate. Regarding TP, both the aerated and non-aerated VF CW units noticeable contributed to TP removal, reaching 72 ± 10% and 82 ± 13%, respectively. Additional TP removal took place in the aerated HF units, reaching 18 ± 12% and 42 ± 25% for HF1 and HF2, respectively.

In the case of WETWINE plant (paper IV) the global removal of COD was generally high (48 – 95%) for the combination of HUSB and hybrid CWs to treat winery WW. The global removal of TSS varied in the range of 90–97%. The average % TSSr value was 93%. This global % TSSr is only affected by HLR so that the % TSSr decreases when HLR is reduced (R2 = 0.67, p =0.01). The WETWINE plant operated at an overall SLR of 81 g COD/m2 ·d higher than that of g 37 COD/m2 ·d applied by Serrano et al. (2011). The higher capacity of the WETWINE plant should be attributed to the larger VF/HF area ratio which equals 1 compared to 0.17 for the Serrano et al. (2011) plant. The optimal COD balance over a 100% influent establishes a 10% COD removal for HUSB, 81% for VF and 5.4% for HF, reaching a total removal 96. % (this is 10%, 80% and 60% CODr for HUSB, VF and HF, respectively, on a step basis). In the case of TSS, the overall balance indicates 67% removal for HUSB, 4.1% for VF and 23% for HF, reaching 94% total removal (67%, 13% and 80% TSSr for HUSB, VF and HF, respectively, on a step basis).

The WETWINE project uses a combination of a HUSB digester and hybrid CWs for effectively treat the winery WW produced along the year, with the consequent variations in flow and load and to produce a final effluent quality.

Hybrid CWs is a treatment train consisting of a combination of different types of CW units (VF and HF) placed sequentially. These units work together to optimize the treatment and removal of TSS, COD, BOD_5 and nitrogen compounds in WW. The FV

CWs played a crucial role in the BOD_5 removal process, allowing the HF CWs to provide a final refinement in the removal of these compounds. This sequential treatment approach, where VF CWs serve as an initial high efficiency step for BOD_5 removal, followed by HF CWs, contributes to the comprehensive treatment of WW. In addition, the nitrification process occurs mostly in FVs due to their aerobic zones. However HF CWs, due to their anaerobic zones, facilitate denitrification processes. By combining these processes in a hybrid system, nitrogen removal efficiency is enhanced, leading to improved overall treatment performance.

The results of this thesis are in line with those published by Brix and Arias (2005). VF CWs can remove COD and BOD_5 as well as retain TSS from WW as nitrifying effectively. In the HF CWs, the treatment of WW moves horizontally through the system substrate, in contact with the plant roots, and rhizomes towards the system outlet (Vymazal, 2009). In this system, the treatment of WW, occurs while water travels below-ground in a water saturated bed and the combination of biological, chemical and physical process as wastewater pass through aerobic, anaerobic, and anoxic zones. (Kadlec and Wallace, 2008; Vymazal, 2014). Oxidized nitrogen compounds are effectively removed in anaerobic and anoxic environments predominating in the HF unit.

6. Conclusions

This thesis discusses the use of CWs in various configurations for treating different types of WW, including urban and industrial WW from food and winery facilities.

The thesis explores the use of a HUSB reactor in combination with aerated CWs for WW treatment. These configurations were found to be effective in treating various types of WW.

CWs are typically considered suitable for treating wastewater effluents up to 2000 people equivalents (pe) due to their land surface requirements. The results obtained in Paper I suggest that the technology could be extended to serve larger populations, particularly in the range of 2000 to 5000 pe. In this range of application, septic tanks are not useful and HUSB digesters can clearly compete with Imhoff tanks and other wastewater pretreatments.

Paper III presents results showing that an aerated VF CWs was successful in treating a significantly higher loading rate, specifically from the food industry, while maintaining treatment efficiency.

In Paper IV, a combination of a HUSB digester and CWs was used to treat winery wastewater. This configuration was evaluated over two years and was found to effectively treat winery wastewater, which often exhibits variations in flow and load. The system produced a final effluent quality suitable for irrigation and helped reduce the carbon footprint of winery operations. The results indicate that the combination of HUSB and CWs is an attractive and robust technology for treating wastewater from moderate-sized wineries. This system effectively meets discharge limits, has low maintenance and operating costs, and produces treated water suitable for agricultural irrigation.

Results provides some answers as to how design parameters and specific may affect results of WW treatment in aerated CWs.

Pre-treatment in HUSB digester and clogging prevention

The combination of HUSB reactor with HF CWs engineered with aeration is indeed an innovative configuration, aimed at enhancing the capacity and effectiveness of CWs while potentially helping to prevent clogging. Clogging is a common challenge in traditional CWs due to the accumulation of organic and inorganic materials over time. The results of paper I, II and IV showed that the introduction of HUSB digester can help pre-treat the urban and industrial (food and winery) WW by breaking down and digesting organic matter, reducing the load of solids and preventing clogging issues in downstream HF CWs.

The HUSB unit removed 76-89 % in urban WW and 67% in food WW of TSS. This unit decreased its performance of TSS removal for winery WW. This was predictable due to the type of WW generated in this type of industry. The winery effluents are constituted by soluble and insoluble phases with low value of pH.

HUSB reactor can also lead to improved removal of organic matter and suspended solids from the influent, reducing the load on the HF CWs. Clogging can thus be minimised and the bed CWs life extended by selecting a HUSB as pre-treatment.

Aeration and surface area reduction

Aeration strategies, depth bed, removal efficiency are factors which affect the surface area required for CWs. The studies of papers I, II, III and IV demonstrate the positive impact of aeration in CWs on treatment efficiency for municipal and industrial (food and winery) WWs and the potential for reducing the required surface area.

Compared to the non-aerated HF unit, aerated units in the HIGHWET pilot plant (Paper I) reached 3.7 times higher BOD_5 average SRRs to treat urban WWs. In KT Food pilot plant BOD_5 percentages above 95% were reached to treat food industrial WWs. In addition, this configuration was able to treat the WW from a winery with an initial load of 600 mg TSS /L and 8000 mg COD/L and obtain a treated effluent with the following characteristics: 35 mg TSS/L and 216 mg COD/L. The combination of HUSB reactor and aerated CWs can enhance the overall treatment efficiency of the system.

The most noteworthy outcome is the potential reduction in the required surface area of the CWs. If aeration can lead to such substantial improvements in treatment efficiency, it means that a smaller area is needed to achieve the same level of treatment. In this thesis, the required area may be reduced by a factor of 5 or more. In addition, from the results one can conclude that the combination of CWs can be adapt to treat the heavy and variable load of wine industry while producing water of suitable for agricultural irrigation. This has significant implications for the practical application of CWs technology.

Aeration regime and recirculation controls nitrogen removal

The thesis discussed the use of intermittent aeration and its effect on pollutant removal efficiency. TN removal varied depending on the operating conditions and was favoured by effluent recirculation. It found that higher ammonia removal rates were achieved with longer aeration periods (5h/3h on/off), while shorter periods (3h/5h on/off) resulted in lower nitrification efficiency. Recirculation was identified as a good option to improve TN removal in aerated CWs.

Adding sorbent materials for phosphorus removal

During this thesis two materials were analysed to remove total phosphorus (TP): The Polonite and the Tobermorite. The Polonite achieved a consistent and relatively high TP removal rate of 56 ± 5% throughout the study. The TP removal rate in the Polonite was maintained at this level, even at a low TP loading rate of 0.2 g $TP/m^2/d$. This material appears to be effective at removing TP from the WW, and its performance remained stable during the study period.

The Tobermorite initially achieved a TP removal rate of about 50%. However, over the course of six months of treatment, the TP removal rate in the Tobermorite decreased to 11%. This decrease in TP removal efficiency in the Tobermorite is noteworthy and suggests a decline in its performance over time.

These findings indicate that the Polonite was more effective at maintaining consistent TP removal rates, even at a low TP loading rate. In contrast, the Tobermorite initially performed well but experienced a decline in TP removal efficiency over the study period. The reasons for this difference in performance may depend on various factors, including the characteristics of the materials, the nature of the WW, and any potential changes in the unit's condition or operation. Further investigation would be needed to understand the underlying causes of the observed differences in TP removal.

Treating urban and industrial wastewater

Highlights the effectiveness and versatility of combining HUSB digesters and aerated CWs for the treatment of various types of WW, with a particular focus on applications in urban and industrial settings, including wineries. The research findings support the potential extension of CWs technology to larger populations and emphasize the attractiveness of this approach for treating moderate-sized winery WW while meeting regulatory requirements.

Hybrid CWs system

The thesis analyzes the use of the combination of HUSB and hybrid CW to treat high load WW such as the water from a winery and the water from a food industry. The results obtained show that this combination is a good alternative for the treatment of these types of WW. This hybrid system is capable of adapting to and withstanding the typical variations in load and flow of a winery. In addition, it also proved to be effective in the treatment of highly loaded WW from the food industry.

In summary, this thesis provides valuable insights into the performance and sustainability of aerated CWs for WW treatment, considering factors such as water depth, aeration regime and pollutant removal efficiency. It highlights the potential benefits of aeration in improving treatment performance and reducing land area requirements, while also emphasising the importance of optimising aeration practices for sustainability. This thesis aims to advance the understanding of how the combinations of HUSB reactor and aerated

CWs can effectively treat urban and industrial WW. By optimising design parameters and considering the specific factors that affect treatment in these systems, this research can contribute to more efficient and sustainable WW treatment practices.

7. Bibliography

Akratos CS, Tsihrintzis VA. (2007). Effect of temperature, HRT, vegetation and porous media on removal efficiency of pilot scale horizontal subsurface flow constructed wetlands. Ecological Engineering. 2007;29:173–191. DOI:10.1016/j.ecoleng.2006.06.013

Allen, W. (2013). The Conservation Fund (referenced in World Resources Institute, Natural Infrastructure – Investing in Forested Landscapes for Source Water Protection in the United States. https://www.wri.org/sites/default/files/wri13_report_4c_naturalinfrastructure_v2.pdf). Accessed 2023-07-28

Álvarez JA, Ruíz I, Soto M. (2008). Anaerobic digesters as a pretreatment for constructed wetlands. Ecological Engineering. 2008;33:54–67. https://doi.org/10.1016/j.ecoleng.2008.02.001

Andoh, B., (2014), Bridging the Gap: Blue Infrastructure Helps Mitigate Rift Between Green, Gray Development. http://www.waterworld.com/articles/print/volume-30/issue-2/urban-water-management/bridging-the-gap-blue-infrastructure-helps-mitigate-rift-between-green-gray-development.html. Accessed 2023-07-24.

Arias C. A., Oirschot D., Kilian R., Pascual A., Carvalho P., Lva T., Zhang Y., Brix H. and Alvarez J.A. (2015). Design and performance evaluation of a highly loaded aerated treatment wetland managing effluents from a food processing industry in Denmark. Water Practice & Technology, 10 (4), 644-651. Doi: 10.2166/wpt.2015.074.

Bodin H., (2013). WW treatment in constructed wetlands: effects of vegetation, hydraulics and data analysis methods. Thesis doctoral Department of Physics, Chemistry and Biology IFM Biology Linköping University.

Brix H. and Johansen N. H. (2004). Guidelines for the Establishment of Reed Bed Systems up to 30 PE (Retningslinier for etablering af beplantede filteranlæg op til 30 PE). Økologisk Byfornyelse og Spildevandsrensning No.52, Miljøstyrelsen. Miljøministeriet, Copenhagen, Denmark (in Danish).

Brix H., Arias C.A. (2005) Danish guidelines for small-scale constructed wetland system for onsite treatment of domestic sewage. Water Science and Technology 51(9):1-9. DOI:10.2166/wst.2005.0275

Carballeira T, Ruiz, I, Soto M. (2016) Effect of plants and surface loading rate on the treatment efficiency of shallow subsurface constructed wetlands. Ecological Engineering. 2016;90:203–214. https://doi.org/10.1016/j.ecoleng.2016.01.038

De la Varga, D., Díaz, A., Ruiz, I., Soto, M., (2013a). Avoiding clogging in constructed wetlands by using anaerobic digesters as pretreatment. Ecological Engineering. 52, 262–269. https://doi.org/10.1016/j.ecoleng.2012.11.005.

De la Varga, D., Ruiz, I., Soto, M., (2013b). Winery wastewater treatment in subsurface constructed wetlands with different bed depths. Water Air Soil Pollution, 224, 1485 – 1497. https//doi.org/10.1007/s11270-013-1485-5.

De la Varga D, Ruiz I, Álvarez JA, Soto M. (2015). Methane and carbon dioxide emissions from constructed wetlands receiving anaerobically pretreated sewage. Science of the Total Environment. 2015;538:824–833. https://doi.org/10.1016/j.scitotenv.2015.08.090

Dolman, N., & O'Donnell, E., (2021), 5 lessons learned from blue-green infrastructure delivery. https://www.ice.org.uk/news-and-insight/the-civil-engineer/july-2021/theory-and-practice-of-blue-green-infrastructure. Accessed 2023-07-24.

Dotro G, Langergraber G., Molle P., Nivala J., Puigagut J., Stein O., von Sperling M., (2017). Biological WW Treatment Series (Volume 7): Treatment Wetlands. https://www.iwapublishing.com/open-access-ebooks/3567. ISBN: 9781780408767. Accessed 2023-07-26

Freeman, A.I., Surridge, B.W.J., Matthews, M., Stewart, M., Haygarth, P.M., (2018). New approaches to enhance pollutant removal in artificially aerated wastewater treatment systems. Science of the Total Environment, 627, pp. 1182-1194. DOI: 10.1016/j.scitotenv.2018.01.261

Fu J., Zhao Y., Yang T., Yao Q., Ji B., Chen S., Dai Y., Tao ., Zha X., (2023). A glance of configuration-operational strategies and intensification of constructed wetland towards land-effective occupation. Journal of Water Process Engineering. Volume 56, December 2023, 104473 https://doi.org/10.1016/j.jwpe.2023.104473.

Ilyas, H., van Hullebusch, E.D.(2020) Performance comparison of different types of constructed wetlands for the removal of pharmaceuticals and their transformation products: a review. Environment Science and Pollution Research 27, 14342–14364 (2020). https://doi.org/10.1007/s11356-020-08165-w.

John, Y., Langergraber, G., Adyel, T.M., Emery David Jr, V., (2020). Aeration intensity simulation in a saturated vertical up-flow constructed wetland. Science of the Total Environment, 708, art. no. 134793. https://doi.org/10.1016/j.scitotenv.2019.134793.

Kadlec, R.H., Wallace, S.D., 2008. Constructed Wetlands, second edition. Taylor & Francis Group, Boca Raton. ISBN 9781420012514, 1420012517

Kadlec R., Wallace S., (2009). Treatment Wetlands (2nd ed.). Boca Raton, FL: CRC Press. ISBN 978-1-56670-526-4 (alk. paper)

Knowles P., Dotro G., Nivala J., García J. (2011) Clogging in subsurface-flow treatment wetlands: Occurrence and contributing factors. Ecological Engineering 37(2):99-112. https://doi.org/10.1016/j.ecoleng.2010.08.005

Laber J., Perfler R., Haberl R. (1997) Two strategies for advanced nitrogen removal in vertical flow constructed wetlands. Water Science and Technology 35(5):71-77. https://doi.org/10.2166/wst.1997.0168

Rossmann M, Teixeira A, Carneiro E, Fonseca F, Carraro A., (2013). Effect of influent aeration on removal of organic matter from coffee processing WW in constructed wetlands. Journal of Environmental Management 2013; 128:912–919. https://doi.org/10.1016/j.jenvman.2013.06.045

Rous V., Vymazal J., Hnátková T., (2019). Treatment wetlands aeration efficiency: A review. Ecological Engineering. https://doi.org/10.1016/j.ecoleng.2019.06.006.

Sánchez M., Ruiz I., Soto M., (2022). The Potential of Constructed Wetland Systems and Photodegradation Processes for the Removal of Emerging Contaminants. A Review, Environments, MDPI. 9 (2022) 116. https://doi.org/10.3390/ENVIRONMENTS9090116

Serrano, L., de la Varga, D., Ruiz, I., Soto, M., (2011). Winery wastewater treatment in a hybrid constructed wetland. Ecological Engineering. 37 (5), 744–753. https://doi.org/10.1016/j.ecoleng.2010.06.038.

Stefanakis Alexandros (2019). The Role of Constructed Wetlands as Green Infrastructure for Sustainable Urban Water Management. Sustainability 2019, 11, 6981; https://doi:10.3390/su11246981.

Štrbac S., Kašanin-Grubin M., Pezo L., Stoji´c N., Loncar B., Curcic L., and Pucarevi´c M., (2023). Green Infrastructure Designed through Nature-Based Solutions for Sustainable Urban Development. International Journal of Environmental Research and Public Health 20, 1102. https://doi.org/10.3390/ ijerph20021102.

Swarnakar A., K., Bajpai S., Ahmad I., (2022). Various Types of Constructed Wetland for WW Treatment- A Review IOP Conference Series: Earth and Environmental Science 1032 (2022) 012026 https://doi:10.1088/1755-1315/1032/1/012026.

Torrens A., (2015). Subsurface flow constructed wetlands for the treatment of WW from different sources. Design and operation. PhD Thesis, Universitat de Barcelona.

V. Torrijos, O.G. Gonzalo, A. Trueba-Santiso, I. Ruiz, M. Soto, Effect of by-pass and effluent recirculation on nitrogen removal in hybrid constructed wetlands for domestic and industrial wastewater treatment, Water Research 103 (2016) 92–100. https://doi.org/10.1016/j.watres.2016.07.028

Trang, N.T.D., Konnerup, D., Schierup, H.H., Chiem, N.H., Tuan, L.A., Brix, H., (2010). Kinetics of pollutant removal from domestic WW in a tropical horizontal subsurface flow constructed wetland system: effects of hydraulic loading rate. Ecological Engineering 36, 527–553. https://doi.org/10.1016/j.ecoleng.2009.11.022

Tunçsiper B, Drizo A, Twohig E. (2015) Constructed wetlands as a potential management practice for cold climate dairy effluent treatment. USA CATENA (135); 2015, pp. 184–192. DO - 10.1016/j.catena.2015.07.028

Vera-Puerto I., Campal J., Martínez S., Cortés-Rico L., Coy H., Tan S., Arias C.A., Baquero-Rodríguez G., and Rosso D., (2022). Effects of Environmental Conditions and Bed Configuration on Oxygen Transfer Efficiency in Aerated Constructed Wetlands. Water 2022, 14, 3284. https://doi.org/10.3390/w14203284.

Vymazal, J. (2007) Removal of nutrients in various types of constructed wetlands. Science of the Total Environment. 380, 48–65 DOI:10.1016/j.scitotenv.2006.09.014

Vymazal J., Kröpfelová L. (2008) WW Treatment in Constructed Wetlands with Horizontal Sub-Surface Flow: Springer. DOI:10.1007/978-1-4020-8580-2

Vymazal J, (2010). Constructed Wetlands for WW Treatment. Water 2, 530-549. https://doi.org/10.3390/w2030530.

Vymazal J. (2013). The use of hybrid constructed wetlands for WW treatment with special attention to nitrogen removal: A review of a recent development. Water research Volumen 47, Issue 14, 15 September 2013. Pages 4795-4811. https://doi.org/10.1016/j.watres.2013.05.029

Vymazal, J., 2014. Constructed wetlands for treatment of industrial wastewaters: a review. Ecological Engineering. 73, 724–751. https://doi.org/10.1016/j.ecoleng.2014.09.03.

Wallace S. D. (2001). Patent: System for removing pollutants from water. United States: US 6,200,469 B1.

Wallace S., and Nivala J., (2008). Statistical Analysis of Treatment Performance in Aerated and Nonaerated Subsurface Flow Constructed Wetlands. 10.1007/978-1-4020-8235-1_15. DOI:10.1007/978-1-4020-8235-1_15

Wallace S.D. (2014). Reducing wetland area requirements by using intensification strategies. In: Proceedings of the 14th IWA Specialist Group Conference on Wetland Systems for Water Pollution Control, 4-8 October 2010. IWA: Shangai, China. pp. 54-68.

Wallace S., Oirschot, D., Stefanakis, A., (2020). Wetland Technology: Practical Information on the Design and Application of Treatment Wetlands - Chapter 5.7 (pp.105-107) Publisher: IWA Publishing.

Wießner, A. & Kuschk, P. & Kaestner, Matthias & Stottmeister, Ulrich. (2002). Abilities of Helophyte Species to Release Oxygen into Rhizospheres with Varying Redox Conditions in Laboratory-Scale Hydroponic Systems. International Journal of Phytoremediation - INT J PHYTOREMEDIAT. 4. 1-15. 10.1080/15226510208500069. DOI:10.1080/15226510208500069

Zhang J., Wu H., Hu Z., Liang S., and Fan J., (2014). Examination of oxygen release from plants in constructed wetlands in different stages of wetland plant life cycle. Environmental Science and Pollution Research volume 21, pages9709–9716 (2014). DOI:10.1007/s11356-014-2905-9

Zhang L., Zhang L., Liu Y., Shen Y., Liu H., Xiong Y. (2010) Effect of limited artificial aeration on constructed wetland treatment of domestic wastewater. Desalination. 2010; 250(3):915–920. DOI:10.1016/j.desal.2008.04.062

Zhang S., Li G., Chang J., Li X., Tao L., (2015). Aerated enhanced treatment of aquaculture effluent by three-stage, subsurface flow constructed wetlands under a high loading rate. Polish Journal of Environmental Studies 2015; 23(5):1821–1830

Zhang Y., Jyu T., Zhang L., Button M., Arias C. A., Weber K., Shi J., Chen Z., Carvalho P., (2018). Microbial community metabolic profiles in saturated constructed wetlands treating iohexol and ibuprofen. Science of The Total Environment Volume 651, Part 2, 15 February 2019, Pages 1926-1934. DOI: 10.1016/j.scitotenv.2018.10.103

Zhao L., Zhu W., Tong W., (2009). Clogging processes caused by biofilm growth and organic particle accumulation in labscale vertical flow constructed wetlands. Journal of Environmental Sciences. 2009;21:750–757. DOI: 10.1016/s1001-0742(08)62336-0

Zapater-Pereyra M, Gashugi E, Rousseau DPL, Alam MR, Bayansan T, Lens PNL (2014). Effect of aeration on pollutants removal, biofilm activity and protozoan abundance in conventional and hybrid horizontal subsurface-flow constructed wetlands. Environmental Technology. 2014;35:2086– 2094. https://doi:10.1080/09593330.2014.893024

8. Annexes

The 4 articles of this thesis are attached as annexes.:

Annexe I: *Hydrolitic anaerobic reactor and aerated constructed wetland systems for municipal WW*

Annexe II: *Aerated Constructed Wetlands for Treatment of Municipal and Food IndustryWW. ConstructedWetlands for IndustrialWW Treatment*

Annexe III: *Horizontal flow aerated constructed wetlands for municipal WW treatment: the influence of bed depth*

Annexe IV: *Nature based solutions for winery WW valorisation*

Annex I

Environmental Technology

ISSN: 0959-3330 (Print) 1479-487X (Online) Journal homepage: http://www.tandfonline.com/loi/tent20

Hydrolytic anaerobic reactor and aerated constructed wetland systems for municipal wastewater treatment – HIGHWET project

A. Pascual, D. de la Varga, C. A. Arias, D. Van Oirschot, R. Kilian, J. A. Álvarez & M. Soto

ENVIRONMENTAL TECHNOLOGY, 2016
http://dx.doi.org/10.1080/09593330.2016.1188995

Taylor & Francis
Taylor & Francis Group

Hydrolytic anaerobic reactor and aerated constructed wetland systems for municipal wastewater treatment – HIGHWET project

A. Pascual[a,b], D. de la Varga[c], C. A. Arias[d], D. Van Oirschot[e], R. Kilian[f], J. A. Álvarez[b] and M. Soto[a]

[a]Department of Physical Chemistry and Chemical Engineering I, University of A Coruña, A Coruña, Galiza, Spain; [b]AIMEN, Porriño – Pontevedra, Spain; [c]Sedaqua (Spin-off from University of A Coruña), O Porriño, Pontevedra, Spain; [d]Department of Bioscience, Aarhus University, Aarhus, Denmark; [e]Rietland bvba, Minderhout, Belgium; [f]Kilian Water, Bryrup, Denmark

ABSTRACT
The HIGHWET project combines the hydrolytic up-flow sludge bed (HUSB) anaerobic digester and constructed wetlands (CWs) with forced aeration for decreasing the footprint and improving effluent quality. The HIGHWET plant in A Coruña (NW of Spain) treating municipal wastewater consists of a HUSB and four parallel subsurface horizontal flow (HF) CWs. HF1, HF2 and HF3 units are fitted with forced aeration, while the control HF4 is not aerated. All the HF units are provided with effluent recirculation, but different heights of gravel bed (0.8 m in HF1 and HF2, and 0.5 m in HF3 and HF4) are implemented. Besides, a tobermorite-enriched material was added in the HF2 unit in order to improve phosphorus removal. The HUSB 76–89% of total suspended solids (TSS) and about 40% of chemical oxygen demand (COD) and biological oxygen demand (BOD). Aerated HF units reached above 96% of TSS, COD and BOD at a surface loading rate of 29–47 g $BOD_5/m^2{\cdot}d$. An aeration regime ranging from 5 h on/3 h off to 3 h on/5 h off was found to be adequate to optimize nitrogen removal, which ranged from 53% to 81%. Average removal rates of 3.4 ± 0.4 g total nitrogen (TN)/$m^2{\cdot}d$ and 12.8 ± 3.7 g TN/$m^3{\cdot}d$ were found in the aerated units, being 5.5 and 4.1 times higher than those of the non-aerated system. The tobermorite-enriched HF2 unit showed a distinct higher phosphate (60–67%) and total phosphorus (54%) removal.

ARTICLE HISTORY
Received 15 February 2016
Accepted 9 May 2016

KEYWORDS
HUSB anaerobic digester; aerated constructed wetlands; nitrogen removal; phosphorus removal; municipal wastewater

1. Introduction

Wastewater treatment of small populations, food and beverage companies, and livestock farms is one of the most important problems due to decentralized location, limited economic resources and lack of specialized personnel. Even though the Water Framework Directive 2000/60/EC, concerning urban wastewater treatment, forces to treat sewage, there are a lot of small and medium-sized towns without this service. In addition, many of them cannot deal with energy and maintenance costs of conventional treatment plants, making them unsustainable and uneconomic.[1]

Constructed wetlands (CWs) are engineered treatment systems for wastewater effluents up to 2000 habitant equivalents, showing a high sustainability potential when properly designed and maintained. CW systems are based on the functioning of natural ecosystems, and the treatment processes involve complex interactions between soil, water, plants and micro-organisms.[2,3] The main parts of a CW are the liner separating the wetland from the subsoil to avoid infiltration and pollution of ground water, substrate bed, vegetation, and influent distribution and effluent collecting systems.

The most used types of CWs are the surface flow or free water surface systems, the horizontal subsurface flow systems (HF) and the vertical subsurface flow systems (VF). For improving the performance and the removal of pollutants and nutrients, a combination of these systems is used, so-called hybrid systems. HF systems are usually operated in conditions of permanent water saturation which limits oxygen transfer and nitrification only occurs at a low rate. Pulsed fed VF systems are partially saturated and can provide good conditions for nitrification, but the denitrification in these systems is limited. Therefore, in hybrid systems, the strengths and weaknesses of each type of system balance each other out and in consequence, it is possible to obtain an effluent low in biological oxygen demand (BOD) and in total nitrogen (TN) concentrations.[3] Many combinations are possible, including subsurface HF followed by VF, VF followed by HF and other stages of filters including water recirculation from one stage to another.[2]

CW technology is able to treat different types of wastewater, and hybrid CW systems can reach

CONTACT M. Soto m.soto@udc.es Department of Physical Chemistry and Chemical Engineering I, University of A Coruña, Rúa da Fraga no. 10, 15008 A Coruña, Galiza, Spain

simultaneous removal of organic matter and nitrogen, but they do not reduce the large surface area demand of these systems. In order to supply the extra oxygen needed to warrant the removal efficiency, while maintaining the advantages provided by CW technology, an external supply of air can be provided.[4–6]. Thus, the concept of aerated CW systems (i.e. with artificial forced aeration) arises.

Forced aeration strategies can vary extensively from partial to total aeration in relation to time and space, and from low to high intensity. The depth of aerated HF CWs varied from 0.3 to 1 m,[6–8] but probably an efficient aeration process requires a high depth in order to reach sufficient long contact time between the supplied air and wastewater. Air pumps can be activated when the oxygen concentration in the units is lower than 0.2 mg/L and turned off when the oxygen concentrations in the CW is higher than 0.6 mg/L.[8] However, this strategy requires automated devices and, usually, continuous aeration was provided, over the overall wetland bed or only near the inlet zone.[6,7] However, a timetable for intermittent aeration or spatial segregation of aerated and non-aerated zones has been considered convenient in order to reach simultaneous nitrification and denitrification.[9–11] In an efficient aerated HF CW, nitrification occurs when the aeration system is turned on, while denitrification requires anoxic conditions which could be obtained by ceasing aeration. Denitrification also requires a carbon source which must be furnished by the influent wastewater substrate. Thus, effluent recirculation could be necessary to improve the contact between the generated nitrate, the influent organic substrate and microbial population during anoxic periods.

Two additional points of interest concerning CW construction and use are clogging prevention and phosphorus removal improvement. One of the main problems with operating subsurface HF systems is clogging of granular media. Although the clogging phenomenon is an extremely complex and not well-understood process, the influent content in suspended solids is an important factor in causing clogging. The use of anaerobic digesters as pretreatment can achieve high suspended solids removal and contribute to avoid wetland clogging problems.[12–15] On the other hand, CWs will be of great interest when combined with anaerobic digesters, as both technologies agree with the essential criteria of low cost and sustainability, including simplicity of construction, operation and maintenance.

Phosphorus removal in CWs can occur through a combination of several processes: plant uptake, microbial growth, adsorption, precipitation within substrates, etc. Among these processes, adsorption and precipitation play the largest role in phosphate removal.[16] So, to ensure efficient phosphorus removal, research should aim to identify substrates that have a high phosphorus removal capacity, and suitable properties for use as CW substrate. Particularly, there is a great interest in studying the beneficial reuse options of by-products and waste materials which can be an alternative to gravel from quarry and which can contribute to the improvement of wastewater treatment systems.

The HIGHWET project was addressed to improve the capacity and effectiveness of CWs as high-rate and sustainable wastewater treatment systems. The HIGHWET project aimed to perform and validate new approaches based on the combination of the hydrolytic up-flow sludge bed (HUSB) anaerobic digester and CWs with forced aeration for decreasing the required surface of conventional HF CWs and improving the final effluent quality. For this purpose, two demonstration plants were designed and constructed in Spain and Denmark. The first configuration (A Coruña, NW of Spain) consisted of a HUSB and HF CWs for raw municipal wastewater treatment, while the second configuration (nearby Aarhus, Denmark) consisted of a combination of a HUSB and hybrid (FV + HF) CWs for the treatment of high-load organic industrial wastewater. The effect of effluent recirculation, aeration regime, energy consumption and different phosphorus adsorbent materials was planned to be checked in both plants. The effect of HF bed depth on aeration and treatment efficiency, which has not been investigated at the moment in a side-by-side comparison at the field scale, was assessed in A Coruña plant. This work reports the first results obtained in the A Coruña HIGHWET plant.

2. Material and methods

2.1. HIGHWET pilot plant in A Coruña

The pilot plant was built at the outdoors of the Science Faculty of the University of A Coruña in A Coruña (Spain) and the start-up was carried out in July 2014. It consisted of a hydrolytic anaerobic digester (HUSB) followed by four horizontal subsurface flow (HF1, HF2, HF3 and HF4) CWs working in parallel and receiving anaerobic pretreated wastewater. Figure 1 presents the diagram of the pilot plant design. HF1, HF2 and HF3 units are fitted with forced aeration, while the HF4 is not aerated in order to be used as a control. All the HF units were provided with effluent recirculation, but different heights of gravel bed were implemented in order to compare the behaviour of the aeration operating at high organic and hydraulic load in different scenarios. The HF beds were planted with common reeds (*Phragmites australis*) at a density of 16 plants/m^2.

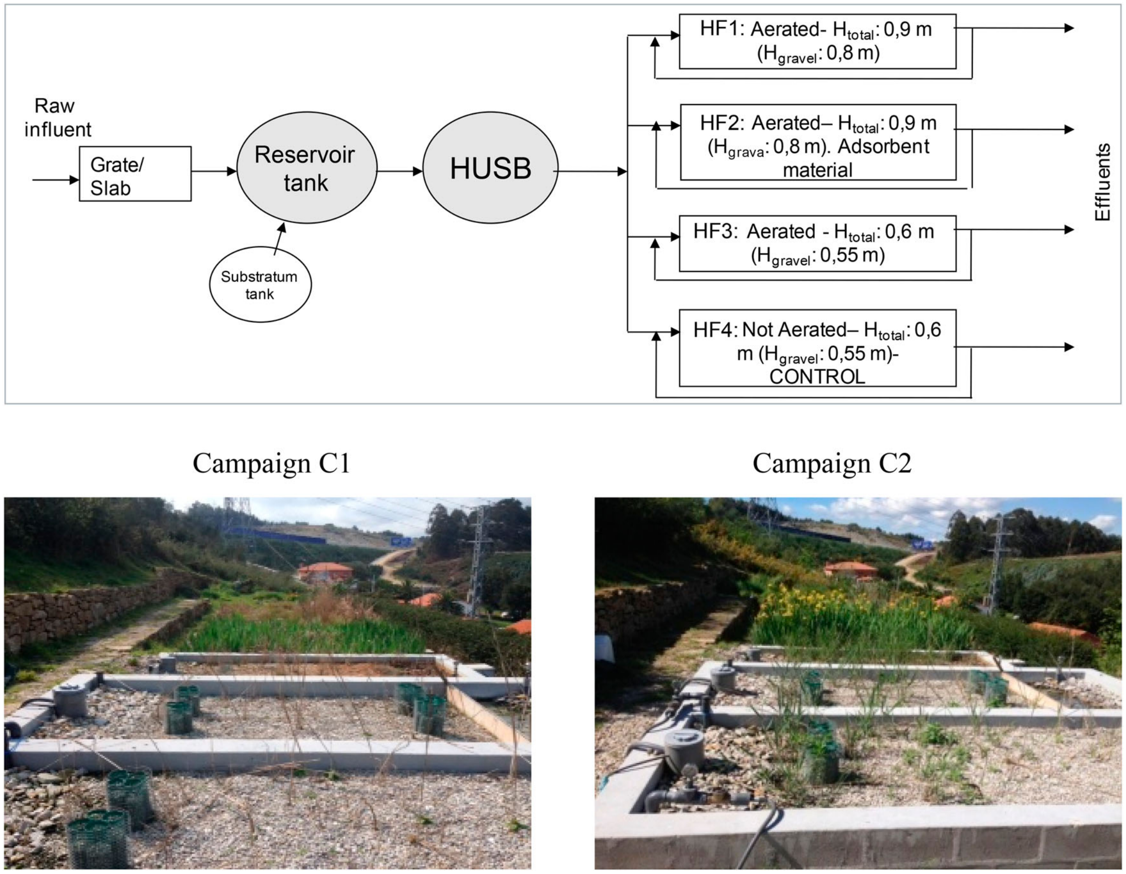


Figure 1. Configuration of the HIGHWET plant at A Coruña and plant situation during campaigns C1 and C2.

The plant was outfitted with the necessary pumps, tanks and other required devices. The raw influent wastewater flowed by gravity to the reservoir tank (volume of 1.8 m^3), after passing the coarse chamber. The coarse chamber was equipped with a mesh of 5 × 10 cm in order to remove big solids and a bypass for draining the excess flow. The concentrated synthetic wastewater was stored in the substratum tank (volume of 600 L) and pumped to the reservoir tank by using a peristaltic pump at a flow rate of 200 L/d. Submerged drain pumps were used in order to stir and homogenize the content of the substratum tank and the reservoir tank. A peristaltic pump at a flow rate of 3000 L/d was used to feed the HUSB reactor, while one three-channel peristaltic pump supplied each aerated bed (HF1, HF2 and HF3) at a flow rate of 1000 L/d and another peristaltic pump supplied the control bed (HF4) at a flow rate of 200 L/d. Finally, four timed submerged drain pumps situated in effluent collection tanks (up to 600 L of capacity) provided the independent recirculation flow to the inlet zone of each one of the HF units. Recirculation pumps operated in ON/OF cycles of 1 h being ON for approximately 10 min (HF1, HF2 and HF3) and 2.5 min (HF4).

2.2. HUSB digester

The HUSB digester consists of a concrete cylinder 0.70 m in diameter and an active height of 1.8 m (active volume of 0.69 m^3 and total height of 2.0 m). It was not provided with a gas/solid/liquid separator, since the target is to operate it at the hydrolytic stage and, therefore, biogas will not be produced or would be kept at a minimum. The design hydraulic retention time (HRT) for the HUSB was 3–7 h, while the organic loading rate (OLR) could be in the range of 0.5–4 g BOD/L·d.

2.3. HF CW units

The four HF units consisted of rectangular basins of 4 × 1.4 m (length × width), giving a total area of 5.6 m^2 for each bed. The base of the beds (with a slope of 1%) and the walls were built in concrete reinforced with iron rods and waterproofed with epoxy painting, in order to secure their isolation. The total height of the basins was 0.9 m for HF1 and HF2 units and 0.6 m for HF3 and HF4 units, while the water plate remained at 0.8 and 0.5 m, respectively. The average porosity was estimated to be 40%; thus, the void volume of each HF unit was 1.8 m^3 for HF1 and HF2 units and 1.1 m^3 for HF3 and HF4 units.

At the inlet zone of each HF unit, the influent distribution system consisted of a pipe (20 cm in diameter and 1.3 m in length) placed perpendicularly to the flow and buried in large gravel (50–60 mm) along the first 50 cm of the bed. This pipe had holes of 30 mm spaced at intervals of 25 cm along the length of the pipe. The effluent collection system was similar to the influent distribution system, but placed at the back of the bed. The water level of each bed was controlled by a pipe elbow inclined to the desired level and placed in an external tank of 40 × 40 × 80 cm. From here, the effluent was driven by a pipeline to the recirculation/final effluent tank.

As the main filtering medium, granitic gravel of 12–16 mm size was implemented for a length of 3 m on HF1 and HF3 units. In the case of the HF2 unit, the granitic gravel (12–16 mm size) was implemented for only a length of 2.5 m, while a phosphorus adsorbent material was placed along the remaining 0.5 m. Both materials in the HF2 unit were separated by perforated plywood. On the other hand, the main filtering medium in the HF4 unit was a finer granitic gravel of 6–12 mm size for a length of 3 m. The height of the filtering media was 0.85 m for HF1 and HF2 units and 0.55 m for HF3 and HF4 units.

2.4. Biomass monitoring in HF units

In order to monitor the development of biomass on the gravel, perforated cylinders of 20 cm diameter were inserted in the gravel, two at 80 cm from the inlet zone of each unit and another two at 80 cm (or at 120 cm in the case of the HF2 unit) from the end of each bed. The perforated cylinders were constructed with a steel mesh with lower mesh size than the gravel diameter and reached the bottom of each bed. Inside each cylinder, four columns of plastic mesh (8 cm in diameter) filled with gravel were placed. The columns will be manually extracted at different times of operation to analyse biomass development and solids accumulation on the gravel surface.

2.5. Aeration system

The aeration system in the HF units consists of a series of pipes installed at the bottom of the beds that will provide the necessary oxygen to the wastewater to maintain the proper concentration of dissolved oxygen (DO) while the water is being treated. The aeration lines are kept pressurized by air pumps that provide uniform distribution of air throughout the bed. The aeration can be adjusted by increasing the aeration time, switching the blowers on and off as the load is increased in the course of the development of the project, in order to adapt to the influent flow and concentration changes.

2.6. Phosphorus adsorbent material

In the HIGHWET project, it will be studied the beneficial reuse options of by-products and waste materials which can be an alternative to gravel or sand from quarry and which can contribute to the improvement of phosphorous removal in CWs. The adsorbent material selected was the scrap material from the production of specific concrete for building material worldwide. It is used for plain masonry or insulating purposes and for reinforced components such as lintels and roof/floor and wall panels.[17] The specific concrete is manufactured from silica sand, cement, lime and water. The material is processed in order to convert the minerals chemically into a strong crystal structure of tobermorite ($Ca_5Si_6O_{16}(OH)_2{\bullet}H_2O$). Tobermorite occurs in nature but is very rare. The adsorbent material based on tobermorite and with a particle size of 0.5–5.6 mm has been implemented in the last 0.5 m of the HF2 unit.

2.7. Influent wastewater

The influent to the plant comes from a local sewer receiving wastewater from one of the faculties of the University of A Coruña and surrounding houses. During the start-up period (July–October 2014), the raw wastewater presented the following characteristics (in mg/L except pH): pH 7.1 ± 0.6, total suspended solids (TSS) 64 ± 47, chemical oxygen demand (COD) 141 ± 87, BOD_5 47 ± 27, NH_4-N 24.2 ± 18.2, TN 25.3 ± 18.3, PO_4-P3.0 ± 2.0. These characteristics corresponded to a much diluted domestic wastewater, which was due to the entrance of rain water in the collection sewer. In order to use a more representative wastewater, in the present study, the raw domestic wastewater was supplemented with a concentrated synthetic substratum. A mixture of substrates (urea, trisodium phosphate, sodium acetate, starch and municipal primary sludge) was stored in the substratum tank (volume of 600 L). The concentrated substratum was formulated and renewed twice a week. This synthetic wastewater was continuously fed to the reservoir tank in order to reach the proposed TSS, BOD and total Kjeldahl nitrogen concentration.

2.8. Sampling and analysis

During the monitoring periods, the pumps were periodically calibrated and corrected to the desired flow, and the actual flow to each unit of the plant was obtained.

Sampling procedures involved taking influent and effluent composite samples (integrated over a 24-h period). Influent samples were collected using an automatic sampler type 1350 of American Sigma, while effluent samples were collected from daily accumulated volume in final tanks. This procedure was repeated once or twice a week. The obtained samples were analysed in the laboratory for total and volatile suspended solids (TSS, VSS), COD, BOD_5, ammonium, TN, nitrate, nitrite, phosphate and total phosphorus (TP). Temperature, pH, oxidation–reduction potential (ORP) and DO were determined *in situ* on the same sampling days. An integrated pH & Redox 26 Crison electrode was used for pH and ORP determination, a selective electrode (Crison 9663) for ammonium and an electrode ProODO from YSI Inc. for DO. Anions and cations were determined by ion chromatography (Metrohm 882/863). Inductively coupled plasma optical emission spectrometry (ICP-OES) was used for TP determination. Analytical methods were carried out as described in Standard Methods.[18]

3. Results and discussion

3.1. Plant operation

Available data for the start-up period (July–December 2014) indicate that the plant operated at an average volumetric loading rate (VLR) of 0.6 g BOD/L·d (HUSB digester) and surface loading rate (SLR) of approximately 20 g BOD_5/m^2 d (aerated HF units) and 4 g BOD_5/m^2 d (non-aerated HF4 unit). A high recirculation flow at the HF units of about 300% of influent flow was applied during this step. In these conditions, the aerated lines completely removed the influent organic matter, reaching TSS, COD and BOD_5 below 7 mg/L (HF1, HF2 and HF3 effluents) and removal rates higher than 98%. The non-aerated line showed slightly lower performance with removal rates of 99% TSS, 93% COD and 89% BOD_5.

Following the start-up period, two detailed monitoring campaigns were carried out in January–February (campaign C1) and March–April (campaign C2) 2015. The operation conditions for these campaigns C1 and C2 are indicated in Table 1 and the influent and effluent characteristics in Table 2. The SLR was increased to approximately 30 (C1) and 40–45 (C2) g BOD_5/m^2 d for the aerated HF units and to 8 (C1) and 14 (C2) for the non-aerated HF unit. On the other hand, the TN SLR remained approximately constant, at 5–6 g TN/m^2·d for the aerated units and 1.5 g TN/m^2·d for the non-aerated unit. In this way, the three aerated HF units received a similar SLR and hydraulic loading rate (HLR), but the HF3 unit operated at a lower HRT (1.2–1.5 d) as well as a higher VLR (166–239 g BOD_5/m^3·d), in comparison to HF1 and HF2 (2–2.9 d and 89–138 g BOD_5/m^3·d). On the other hand, the higher HRT (5.0–5.2 d) corresponded to the non-aerated HF4 unit. The later are typical values of conventional HF systems treating domestic wastewater. The recirculation rate ranged from 110% to 160% of influent flow for the aerated units and from 350% to 370% for the HF4 unit, substantially increasing the actual hydraulic load through the HF beds. An aeration regime of 5 h on followed by 3 h off (overall cycle of 8 h) was set at HF1, HF2 and HF3 units during campaign C1. The aeration rate was decreased to 3 h on/5 h off during campaign C2. During both campaigns, the HUSB operated at the design HRT and OLR.

The influent temperature was approximately 14°C (C1) and 19°C (C2) that decreased to about 11°C (C1) and 15.5°C (C2) at the outlet of the HF units. The DO content was low in the HUSB effluent (1.6–2.3 mg/L) and significantly increased at the outlet of the HF units. DO reached saturation levels at HF1, HF2 and HF3 units at campaign C1. At campaign C2, DO in HF1 and HF2 remained high (7–9 mg O_2/L), while the mean value for the HF3 unit was lower (3.7 mg O_2/L). This behaviour could be related to the higher SLR in the HF3 unit during campaign C2 (47 g BOD_5/m^2·d) in comparison to campaign C1 (33 g BOD_5/m^2·d) and the lower bed depth of the HF3 in comparison to HF1 and HF2 units. The lower bed depth of the HF3 unit could reduce the oxygen transfer efficiency whose effects would only be noticeable in overload conditions. On the other hand, the non-aerated unit HF4 showed moderate OD levels in the range of 4–5 mg/L. Except for the HF2 unit effluent, the pH was very stable throughout the different measurement points in the plant, with mean values ranging from 7.2 to 7.7. The HF2 unit provided with the tobermorite-enriched material showed a distinct increase in pH at the effluent, showing mean values of 8.2 (C1) and 8.3 (C2), significantly higher ($p < .01$) than those of the other HF units.

3.2. HUSB digester performance

An anaerobic digester with a similar design to the typical up-flow anaerobic sludge blanket (UASB) when it is used at hydrolytic (non-methanogenic) conditions is known as a HUSB digester.[19,20] The type of substrate, influent concentration, temperature, HRT and solid retention time are the main operational parameters that define the methanogenic or non-methanogenic conditions of an anaerobic system.[19] The HUSB reactor should minimize methanogenic activity while enhancing hydrolysis and acidification of particulate matter in order to reach high volatile fatty acid concentrations in the effluent. Therefore, a HUSB reactor is mainly used for that wastewater with a high suspended solid concentration in order

Table 1. Hydraulic and loading rate parameters applied at the HIGHWET plant treating municipal wastewater.

Parameter	HUSB	HF1	HF2	HF3	HF4
Campaign C1					
Feeding flow (L/d)	2724	879	789	897	213
HLR (L/m^2·d)	–	157.0	140.9	160.2	38.0
SLR (g BOD$_5$/m^2·d)	–	31.9	28.6	32.6	7.7
SLR (g TN/m^2·d)	–	6.1	5.5	6.2	1.5
VLR (g BOD$_5$/m^3·d) [a]	2116	99	89	166	39
HRT (d)	0.25	2.0	2.3	1.2	5.2
Recirculation flow (L/d)	0.0	1059	1014	1000	789
HLR with recirculation (L/m^2·d)	–	346.1	322.0	338.8	178.9
Aeration regime (h on/ h off)	–	5/3	5/3	5/3	0/24
Campaign C2					
Feeding flow (L/d)	2576	707	631	748	221
HLR (L/m^2·d)		126.3	112.7	133.6	39.5
SLR (g BOD$_5$/m^2·d)	–	44.3	39.6	46.9	13.9
SLR (g TN/m^2·d)	–	5.3	4.7	5.6	1.6
VLR (g BOD$_5$/m^3·d)[a]	2005	138	123	239	71
HRT (d)	0.27	2.5	2.9	1.5	5.0
Recirculation flow (L/d)	0	1059	1014	1000	789
HLR with recirculation (L/m^2·d)	–	315.4	293.8	312.1	180.4
Aeration regime (h on/ h off)	–	3/5	3/5	3/5	0/24

[a]VLR (volumetric loading rate) considering the active volume of the HUSB digester and the void volume of the HF units (see text).

to solubilize particulate matter and to increase the removal of easily biodegradable organic matter in the wastewater.

Treating municipal wastewater, the HUSB digester can operate at HRT ranging from 3 to 14 h.[19,13] In the HIGHWET plant, the HUSB was operated at approximately 6 h of HRT. As shown in Figure 2, the wastewater parameter most affected by the anaerobic treatment is the particulate organic matter. The HUSB removed 76–89% of TSS, highly reducing the influent concentration of suspended solids to the HF units. Average COD and BOD removal was 42% and 48%, respectively. Additionally, the HUSB digester removed organic nitrogen (Org. N, calculated as the difference between TN and ammonia and nitric nitrogen) at a rate ranging from 14% to 22% (Figure 2) and increased the ammonia concentration by about 35%, as can be obtained from Table 2.

During the reported period of 4-month operation, the HUSB did not require sludge purge. Available data from periodic inspections of the height and concentration of the sludge bed indicate a rate of sludge formation of

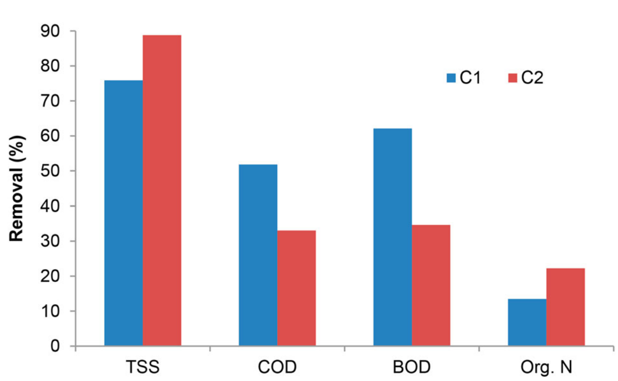


Figure 2. Efficiency of the HUSB digester in removing suspended and total organic matter and Org. N at campaigns C1 and C2.

approximately 2 L/d with a concentration of about 30 g VSS/L and 60 g COD/L. A COD balance indicates that the sludge bed accumulates about 6% of influent COD, which in turn is about 14% of the removed COD. Once the maximum sludge bed of about 1.2 m is reached, maintaining the steady state will require the purge of about half (or more) of the accumulated sludge at each three-month period. The purge frequency could be reduced if a larger HRT or a lower VLR is applied. However, in the case of large plants, the optimum solution may be a more intensive process and a higher purge frequency.

3.3. Organic matter removal in HF units

Instead of the increase in SLR during campaigns C1 and C2 in comparison to the start-up period, the aerated lines completely removed the influent organic matter reaching very low TSS and BOD$_5$ (usually close to zero or below the detection limit) and COD below 23 mg/L (Table 2). Removal percentages higher than 96% were usually achieved. Only the non-aerated HF unit showed slightly lower performance with removal rates of 98% TSS, 86% COD and 91% BOD$_5$ during campaign C1. Thus, all the units showed sufficient capacity to remove the higher SLR applied, which ranged from 29 to 47 g BOD$_5$/m^2·d (50–63 g COD/m^2·d) for the aerated units and from 8 to 14 g BOD$_5$/m^2·d (14–19 g COD/m^2·d) for the non-aerated unit. Average surface removal rates (SRRs) were 37 and 10 g BOD$_5$/m^2·d for the aerated and non-aerated units, respectively.

3.4. Nitrogen removal in HF units

Figure 3 shows the removal of and increase in several forms of nitrogen in the HF units. Nitrogen in the influent

Table 2. Characteristics of the influent and effluent from each unit of the HIGHWET plant treating municipal wastewater

	T	pH	DO	TSS	COD	BOD	TN	NH_4-N	NO_3-N	NO_2-N	PO_4-P	TP
Campaign C1 (*n* = 6)												
Storage tank[a]	12.1 ± 3.8	7.6 ± 0.9	3.2 ± 1.2	568 ± 13.9	739 ± 33	536 ± 67	33.5 ± 14.0	20.8 ± 12.2	0.1 ± 0.2	0.14 ± 0.24	11.3 ± 3.8	nd
HUSB effluent	14.2 ± 1.3	7.3 ± 0.9	2.3 ± 0.8	137 ± 7.4	356 ± 16	203 ± 25	39.0 ± 19.7	27.9 ± 14.4	0.1 ± 0.2	0.22 ± 0.31	11.6 ± 5.9	nd
HF1 effluent	10.9 ± 2.8	7.4 ± 0.5	11.2 ± 0.6	bl	11 ± 5	1.8 ± 3	18.3 ± 8.1	0.0 ± 0.1	13.8 ± 6.7	0.17 ± 0.21	12.0 ± 3.6	nd
HF2 effluent	10.7 ± 2.9	8.2 ± 0.3	10.5 ± 0.9	bl	12 ± 3	1 ± 2	17.5 ± 4.8	3.5 ± 4.3	10.5 ± 2.5	2.12 ± 1.39	4.7 ± 1.5	nd
HF3 effluent	10.8 ± 2.7	7.5 ± 0.4	10.5 ± 1.0	bl	12 ± 5	0.8 ± 0	15.3 ± 7.3	bl	11.4 ± 5.4	0.25 ± 0.19	11.8 ± 3.9	nd
HF4 effluent	10.8 ± 2.5	7.4 ± 0.3	4.9 ± 1.4	3 ± 5	50 ± 29	18.9 ± 2	25.2 ± 3.5	19.8 ± 3.4	0.2 ± 0.3	0.27 ± 0.29	9.5 ± 1.8	nd
Campaign C2 (*n* = 7)												
Storage tank[a]	16.3 ± 2.2	7.2 ± 0.4	1.7 ± 0.6	634 ± 15	699 ± 31	537 ± 67	34.3 ± 8.8	26.1 ± 9.5	bl	0.06 ± 0.17	12.7 ± 2.4	18.6 ± 3.3
HUSB effluent	19.0 ± 1.8	7.5 ± 0.3	1.6 ± 0.6	71 ± 6	468 ± 21	351 ± 44	41.6 ± 11.8	35.2 ± 10.2	bl	0.07 ± 0.17	15.3 ± 2.5	18.3 ± 8.8
HF1 effluent	15.6 ± 1.8	7.6 ± 0.1	6.8 ± 2.4	bl	9 ± 19	bl	13.6 ± 6.0	6.4 ± 7.4	4.2 ± 2.8	0.43 ± 0.32	12.5 ± 2.8	18.8 ± 3.0
HF2 effluent	15.5 ± 1.9	8.3 ± 0.5	9.2 ± 0.9	bl	4 ± 6	bl	8.0 ± 2.9	0.3 ± 0.6	6.2 ± 2.3	0.16 ± 0.26	5.0 ± 1.1	8.5 ± 2.2
HF3 effluent	15.3 ± 1.8	7.7 ± 0.2	3.7 ± 1.3	bl	23 ± 39	bl	19.7 ± 9.0	15.3 ± 8.6	0.6 ± 0.7	0.43 ± 0.39	13.6 ± 2.6	21.6 ± 2.5
HF4 effluent	15.0 ± 1.7	7.6 ± 0.1	4.6 ± 1.1	bl	6 ± 10	bl	23.9 ± 2.2	20.8 ± 2.5	0.2 ± 0.2	0.14 ± 0.19	11.8 ± 1.3	17.2 ± 1.6

[a] Wastewater in the storage tank was the sum of raw wastewater and supplemented synthetic wastewater and was used as the influent to the HUSB digester. bl, below detection limit; nd, not determined. Concentrations are in mg/L.

to the HF units (i.e. HUSB effluent) appeared mainly as ammonia nitrogen (71% and 85% of TN in campaigns C1 and C2, respectively), the remaining corresponding to Org. N (15–28%), while nitric (NO_3^- and NO_2^-) nitrogen accounted for less than 1% of TN. All the aerated units effectively removed TN, Org. N and NH_4^+-N at rates ranging from 47% to 100%, but differences existed mainly as a function of the unit configuration and the on/off aeration ratio.

At the higher aeration ratio (campaign C1, 5 h on/3 h off), units HF1 and HF3 completely nitrified the ammonium, which did not appear in the final effluent. A higher accumulation of NO_3-N in the deeper HF1 unit than in the HF3 unit led to a slightly higher TN removal in HF3 (61%) than in HF1 (53%). The tobermorite-enriched HF2 unit presented during campaign C1 a performance similar to that of the other aerated units, with the only difference of a slightly lower ammonia removal rate (88%) which appeared in the effluent at low concentrations. This could be due to the higher pH in this unit, probably caused by the presence of the tobermorite material that could induce partial inhibition of ammonia nitrification. In the conditions of aeration of campaign C1, the unremoved nitrogen accumulated in the effluent in the form of nitrate, indicating that denitrification was the limiting step. The non-aerated HF unit reached a much lower ammonia removal (29%) and TN (35%), accumulating ammonia but not nitrate in the effluent.

During campaign C2, the aeration regime in HF1, HF2 and HF3 units was changed to 3 h on/5 h off, thus receiving a 40% less air flow. At the same time, the organic SLR increased by about 40% and the nitrogen SLR remained nearly the same or slightly decreased. In comparison to campaign C1, during campaign C2 the percentage ammonia removal decreased in units HF1 and HF3, while it increased in the HF2 unit (Figure 3). TN removal increased in HF1 (67%) and HF2 (81%), while it decreased in HF3 (53%). The better operation of the HF2 unit during campaign C2 could be due to a higher surface area provided by the tobermorite material, once the system adapted to the effect of higher pH. Ammonia accumulated in the effluents of HF1 and mainly of HF3, but not in HF2 effluent. Thus, nitrification became the limiting step in HF1 and HF3 units. Instead of the higher BOD SLR in the non-aerated unit during campaign C2, the percentage of ammonia and TN removal slightly increased up to 41% and 43%, respectively.

A similar percentage removal of Org. N was observed in the aerated HF1 and HF3 units and in the non-aerated HF4 unit, which ranged from 47% to 66%, while the tobermorite-enriched HF2 unit reached distinctly high values of 79–87% (Figure 3). Considering that the Org.

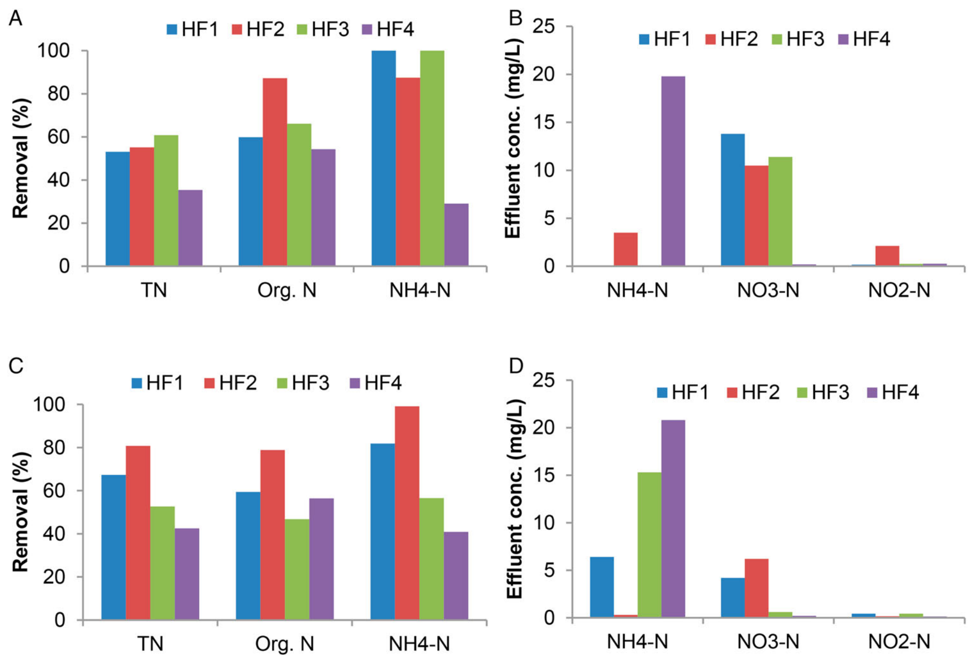


Figure 3. Percentage removal and effluent concentration for several forms of nitrogen in HF units during campaigns C1 (a and b) and C2 (c and d).

N can be removed throughout ammonification, the maximum surface nitrification rate (SNR) can be calculated. The results for maximum nitrification rate and TN removal rate are presented in Table 3. Higher nitrification rates were obtained in campaign C1, clearly decreasing in campaign C2 due to the reduction in aeration flow, particularly in the shallower unit HF3. However, TN removal rate slightly increased in HF1 and HF2 units from campaign C1 to C2. On the other hand, the volumetric nitrification rate (VNR) and particularly the volumetric TN removal rate remained higher in the HF3 unit in both campaigns.

The results in Figure 3 and Table 3 indicated that an aeration regime ranging from 5 h on/3 h off to 3 h on/5 h off may be adequate to optimize TN removal in HF CWs with forced aeration. Thus, a fifty/fifty on/off regime of aeration is recommended as the reference value. As the minimum HRT applied in the present study was 1.2 d, an aeration cycle of 8 h (on + off) may be suitable, but it may need optimization for the operation with diluted wastewater at a shorter HRT. On the other hand, aerated HF systems reached average SRR of 3.4 ± 0.4 g TN/m^2·d and volumetric removal rate (VRR) of 12.8 ± 3.7 g TN/m^3·d, which were 5.5 and 4.1 times higher than those found in the non-aerated system.

3.5. Phosphorus removal

Influent and effluent concentrations of phosphate (campaigns C1 and C2) and TP (C2) are given in Table 2, while Figure 4 shows the percentage removal for the different HF units. The HUSB digester showed a minor effect on TP, but slightly increased phosphate content in the effluent. Reduced phosphate (c. 20%) and TP (6%) removal was obtained in the non-aerated HF unit. These reduced removals are not surprising as the HF4 unit was operated with a SLR (8–14 g BOD$_5$/m^2·d) higher than the recommended design values of 5–6 and because the monitoring campaigns were carried out in winter (C1) and early spring (C2) when the planted macrophyte was still dormant. A lower effect on phosphate and TP was even found in the aerated HF1 and HF3 units, as shown in Figure 4. On the contrary, the tobermorite-enriched HF2 unit reached distinctly high values of

Table 3. Surface and volumetric nitrification and TN removal rates for HF units.

	Campaign C1 (5 h on/3 h off)				Campaign C2 (3 h on/5 h off)			
	HF1	HF2	HF3	HF4	HF1	HF2	HF3	HF4
SNR (g N/m^2·d)[a]	5.4	4.8	5.6	0.5	4.1	4.5	3.1	0.7
VNR (g N/m^3·d)[a]	16.8	14.8	28.6	2.7	12.8	14.0	15.5	3.6
SRR (g TN/m^2·d)	3.2	3.0	3.8	0.5	3.5	3.8	2.9	0.7
VRR (g TN/m^3·d)	10.1	9.4	19.3	2.7	11.0	11.8	14.9	3.6

[a]Maximum nitrification rate by accounting both the removed ammonia and the removed Org. N.

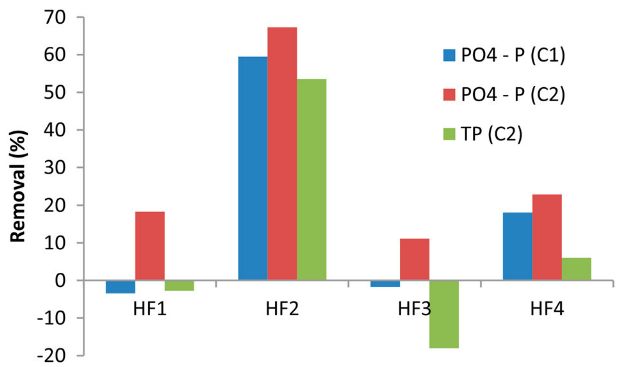


Figure 4. Percentage removal of phosphate and TP in HF units.

60–67% phosphate removal and 54% TP removal. More research is still needed to assess the sustainability of phosphorus removal by this material (lab absorption assays and long-term continuous operation) and particularly the arrangement used to place the tobermorite material in this HIGHWET plant treating domestic wastewater.

3.6. Final remarks

As extensive and passive treatment systems, CWs usually require a high footprint in the form of land surface, thus being considered suitable for the treatment of wastewater effluents below 2000 people equivalents (pe). The SLR generally recommended for secondary treatment in HF CWs is in the range of 4–6 g BOD/m^2·d.[21,22] So, the surface land required ranges from 5 to 7 m^2/pe. In these conditions, HF CWs reach current European Union (EU) targets for secondary treatment, together with a substantial nitrogen and phosphorus removal.[3,22,23] A higher SLR can be applied in HF CWs but in detriment of effluent quality and system sustainability.[13] Vymazal [3] reported that HF CWs ($n = 213$ systems) receiving a SLR of 9.7 g BOD/m^2·d reached an average effluent BOD_5 of 32 mg/L, which is above the EU effluent standards. Furthermore, these systems removed about 0.83 ± 6.56 g TN/m^2·d.[24]

The non-aerated control unit in the present study removed 0.5–0.7 g TN/m^2·d, while it received a SLR ranging from 8 to 14 g BOD_5/m^2·d, thus being comparable to the literature performance of HF systems. Compared to the non-aerated HF unit, aerated units in the present study reached 3.7 and 5.5 times higher BOD_5 and TN SRRs, somewhat higher than those recently reported by Zapater-Pereyra et al. [25] for aerated HF systems. Even the BOD_5 SRR in the aerated units was about eight times higher than the current design criteria for conventional HF units. Thus, we can conclude that the required area can be reduced by a factor of 5, which fulfils the aims of the HIGHWET project. These results could favour the extension of CW technology to serve wastewater discharges in a broad range above 2000 pe, namely in the range of populations from 2000 up to 5000 pe. In this range of application, septic tanks are not useful and HUSB digesters can clearly compete with Imhoff tanks and other wastewater pretreatments.

Three additional points of interest about the performance of HIGHWET plants which are currently being assessed are those related to substrate clogging, greenhouse gases (GHG) and energy consumption. In the present study, the plant included an anaerobic pretreatment step in order to reduce the entrance of suspended solids to HF beds and help in clogging prevention. Additionally, compared to conventional HF units, larger gravel was used in aerated units. We have taken into consideration that clogging can also be caused by biofilm development,[26] which is a risk in more intensive systems. However, clogging processes usually lasted in time and then were not assessed in this first report. On the other hand, CWs can cause an important flux of GHG, such as nitrous oxide (N_2O), carbon dioxide (CO_2) and methane (CH_4), that could mitigate the environmental benefits of CWs. Methane emissions are higher when anaerobic conditions predominate over aerobic conditions, as occurs in conventional HF CWs.[27] Artificial aeration increases aerobic conditions and thus probably reduces CH_4 emissions.[7,28] Reduction in N_2O has been also found in some operation conditions when aeration was provided.[7] Finally, energy consumption due to forced aeration must be assessed taking into account both economic issues and its contribution to indirect GHG emissions.

4. Conclusions

The HIGHWET project combines the HUSB digester and HF CWs with forced aeration for decreasing the footprint and improving the effluent quality. The HUSB removed 76–89% of TSS, highly reducing the influent concentration of suspended solids to the HF units. The average COD and BOD removal was 42% and 48%, respectively, while surplus sludge was produced at a rate equivalent to 6% of influent COD.

HF units showed TSS, COD and BOD removal percentages higher than 96% (aerated units) and 90% (non-aerated unit) when the SLR applied ranged from 29 to 47 g BOD_5/m^2·d (50–63 g COD/m^2·d) for the aerated units and from 8 to 14 g BOD_5/m^2·d (14–19 g COD/m^2·d) for the non-aerated unit. The results obtained indicated that an aeration regime ranging from 5 h on/3 h off to 3 h on/5 h off may be adequate to optimize TN removal in HF CWs with forced aeration. In these conditions, TN removal ranged from 53% to 81%.A fifty/fifty on/off regime of aeration is recommended as the reference value. On the other hand, aerated HF systems reached average SRRs of 3.4 ± 0.4 g TN/m^2·d and VRRs of 12.8 ± 3.7 g TN/m^3·d, which were 5.5 and 4.1 times higher than those found in the non-aerated system.

A reduced phosphate (20%) and TP (6%) removal was obtained in the non-aerated HF unit. A lower effect on phosphate and TP was even found in the aerated HF1 and HF3 units. On the contrary, the tobermorite-enriched HF2 unit reached distinctly high values of 60–67% phosphate removal and 54% TP removal, being a promising approach to increased phosphorus removal in CWs.

Compared to the non-aerated HF unit, aerated units in the present study reached 3.7 and 5.5 times higher BOD_5 and TN SRRs. Even the BOD_5 SRR in the aerated units was about eight times higher than the current design criteria for conventional HF units. In conclusion, the required area may be reduced by a factor of 5 or more, which favours the extension of the CW technology to serve wastewater discharges in a broad range above 2000 pe. Three additional points of interest about the performance and sustainability of the HIGHWET plants which are currently being assessed are those related to substrate clogging, GHG and energy consumption.

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding information

This work was supported by the EU's seventh framework programme for research, technological development and demonstration under grant agreement no. [605445].

References

[1] EC. 6th commission summary on the implementation of the urban waste water treatment directive. Commission Staff Working Paper; 2011.

[2] Brix H, Arias CA. The use of vertical flow constructed wetlands for on-site treatment of domestic wastewater: new Danish guidelines. Ecol Eng. 2005;25:491–500.

[3] Vymazal J. The use constructed wetlands with horizontal sub-surface flow for various types of wastewater. Ecol Eng. 2009;35:1–17.

[4] Wallace S, Liner M, Diebold J. 2007. Degradation of aircraft deicing runoff in aerated engineered wetlands. Multi-functions of wetlands systems, June 26–29, University of Padova, Italy.

[5] Cottingham PD, Davies TH, Hart BT. Aeration to promote nitrification in constructed wetlands. Environ Technol. 1999;20:69–75. doi:10.1080/09593332008616794

[6] Nivala J, Wallace S, Headley T, et al. Oxygen transfer and consumption in subsurface flow treatment wetlands. Ecol Eng. 2013;61:544–554.

[7] Maltais-Landry G, Maranger R, Brisson J. Effect of artificial aeration and macrophyte species on nitrogen cycling and gas flux in constructed wetlands. Ecol Eng. 2009;35:221–229.

[8] Zhang L, Zhang L, Liu Y, Shen Y, Liu H, Xiong Y. Effect of limited artificial aeration on constructed wetland treatment of domestic wastewater. Desalination. 2010;250:915–920.

[9] Maltais-Landry G, Chazarenc F, Comeau Y, Troesch S, Brisson J. Effects of artificial aeration, macrophyte species, and loading rate on removal efficiency in constructed wetland mesocosms treating fish farm wastewater. J Environ Eng Sci. 2007;6:409–414.

[10] Tao W, Wang J. Effects of vegetation, limestone and aeration on nitritation, anammox and denitrification in wetland treatment systems. Ecol Eng. 2009;35:836–842.

[11] Araya F, Vera I, Sáez K, Vidal G. Effects of aeration and natural zeolite on ammonium removal during the treatment of sewage by mesocosm-scale constructed wetlands. Environ Technol. 2016;37:1811–1820.

[12] Ruiz I, Álvarez JA, Díaz MA, Serrano L, Soto M. Municipal wastewater treatment in an anaerobic digester-constructed wetland system. Environ Technol. 2008;29:1249–1256.

[13] Ruiz I, Díaz MA, Crujeiras B, García J, Soto M. Solids hydrolysis and accumulation in a hybrid anaerobic digester-constructed wetlands system. Ecol Eng. 2010;36:1007–1016.

[14] Pedescoll A, Corzo A, Álvarez E, García J, Puigagut J. The effect of primary treatment and flow regime on clogging development in horizontal subsurface flow constructed wetlands: an experimental evaluation. Water Res. 2011;45:3579–3589.

[15] de la Varga D, Díaz A, Ruiz I, Soto M. Avoiding clogging in constructed wetlands by using anaerobic digesters as pre-treatment. Ecol Eng. 2013;52:262–269.

[16] Zhao YQ, Babatunde AO, Hu YS, Kumar JLG, Zhao XH. Pilot field-scale demonstration of a novel alum sludge-based constructed wetland system for enhanced wastewater treatment. Pro Biochem. 2011;46:278–283.

[17] Hellers BG, Schmidt BR. 2011. Autoclaved aerated concrete (AAC) – the story of a low-weight material. 5th International conference on autoclaved aerated concrete, Bydgoszcz, Poland, http://www.5icaac.utp.edu.pl

[18] APHA, 2005. Standard methods for the examination of water and wastewater, 21st ed. American Public Health Association/American Water Works Association/Water Environment Federation, Washington.

[19] Álvarez JA, Zapico CA, Gómez M, Presas J, Soto M. Anaerobic hydrolysis of a municipal wastewater in a pilot-scale digester. Water Sci Technol. 2003;47:223–230.

[20] Álvarez JA, Ruíz I, Soto M. Anaerobic digesters as a pre-treatment for constructed wetlands. Ecol Eng. 2008;33:54–67.

[21] Akratos CS, Tsihrintzis VA. Effect of temperature, HRT, vegetation and porous media on removal efficiency of pilot-scale horizontal subsurface flow constructed wetlands. Ecol Eng. 2007;29:173–191.

[22] Carballeira T, Ruiz, I, Soto M. Effect of plants and surface loading rate on the treatment efficiency of shallow subsurface constructed wetlands. Ecol Eng. 2016;90:203–214.
[23] Barros P, Ruiz I, Soto M. Performance of an anaerobic digester-wetland system for a small community. Ecol Eng. 2008;33:142–149.
[24] Vymazal J. The use of hybrid constructed wetlands for wastewater treatment with special attention to nitrogen removal: a review of a recent development. Water Res. 2013;47:4795–4811.
[25] Zapater-Pereyra M, Gashugi E, Rousseau DPL, Alam MR, Bayansan T, Lens PNL. Effect of aeration on pollutants removal, biofilm activity and protozoan abundance in conventional and hybrid horizontal subsurface-flow constructed wetlands. Environ Technol. 2014;35:2086–2094. doi:10.1080/09593330.2014.893024
[26] Zhao L, Zhu W, Tong W. Clogging processes caused by biofilm growth and organic particle accumulation in lab-scale vertical flow constructed wetlands. J Environ Sci. 2009;21:750–757.
[27] de la Varga D, Ruiz I, Álvarez JA, Soto M. Methane and carbon dioxide emissions from constructed wetlands receiving anaerobically pretreated sewage. Sci Tot Environ. 2015;538:824–833.
[28] Maltais-Landry G, Maranger R, Brisson J, Chazarenc F. Greenhouse gas production and efficiency of planted and artificially aerated constructed wetlands. Environ Pollut. 2009;157:748–754.

Annex II

Part II

Food and Beverage Industry

3

Aerated Constructed Wetlands for Treatment of Municipal and Food Industry Wastewater

A. Pascual[1,3], D. De la Varga[2,3], M. Soto[2,3], D. Van Oirschot[4], R.M. Kilian[5], J.A. Álvarez[1], P. Carvalho[6], H. Brix[6] and C.A. Arias[6]

[1] *AIMEN, Spain*
[2] *Sedaqua (Spin-off from University of A Coruña), Spain*
[3] *University of A Coruña, Spain*
[4] *Rietland bvba, Van Aertselaerstraat 70, Minderhout, Belgium*
[5] *Kilian Water Ltd., Denmark*
[6] *Department of Bioscience – Aquatic Biology, Aarhus University, Aarhus, Denmark*

3.1 Introduction

Constructed Wetlands (CWs) are engineered wastewater treatment systems that have been designed and constructed to mimic processes that occur in natural wetlands. Vegetation, soils, and their associated microbial assemblages are combined to effectively treat wastewater [1].

CWs are shallow basins, generally from 0.3 to 1.0 m. Wastewater can circulate freely, like natural ponds, and this kind of CW is called a free water surface (FWS) system, with aquatic vegetation rooted in the bottom, or floating plants. Another type of CW are planted beds filled with sand or gravel, and they are called subsurface flow systems (SSF). Depending on the flow direction, they are horizontal flow (HF) or vertical flow (VF) systems.

HF are permanently flooded, water flows horizontally and is not exposed to the atmosphere level as it is maintained under the surface (about 1–5 cm). On the other hand, VF wetlands are intermittently pulse-loaded, on top, and wastewater percolates through the unsaturated substrate. Aeration pipes connecting the atmosphere to a manifold of perforated drainage pipes are installed to provide a pathway for air to be drawn into the substrate from the bottom of the bed. Thus, air enters the bed from either the top or the bottom and maintains aerobic conditions in the bed. This approach provides a significant improvement of subsurface oxygen availability compared to HF designs.

Engineered treatment wetlands are other options of CWs systems that might include "reciprocating", also known as "tidal flow" or "fill-and-drain" wetlands. As the wetland bed is drained, air is drawn into the bed [2], oxygenating the exposed biofilms on the wetland substratum. This improves the treatment performance compared to systems with a static water lever [3, 4]. Mechanical aeration of SSF wetlands using air distribution pipes installed at the bottom of the wetland bed has also been utilized as a means to increase oxygen transfer in wetland treatment systems. They are called (artificially) aerated wetlands or Forced Bed Aeration Wetlands (FBA®).

Constructed Wetlands for Industrial Wastewater Treatment, First Edition. Edited by Alexandros I. Stefanakis.

CWs have been used for wastewater treatment for more than fifty years to treat different types of polluted waters around the world. CWs became a widely accepted technology to deal with both point and non-point sources of water pollution as they offer a technical, low-energy, and low-operational-requirements alternative to conventional treatment systems, besides being able to meet discharge standards. Used initially to treat municipal wastewaters, the application of CWs has been expanded to the treatment of industrial effluents, agricultural wastewaters, livestock farm effluents, landfill leachate and stormwater runoff, among others [5–7].

The processes involved in pollutant removal include sedimentation, sorption, precipitation, evapotranspiration, volatilization, photodegradation, diffusion, plant uptake, and microbial degradation processes such as nitrification, denitrification, sulphate reduction, carbon metabolization, among others [8].

CW systems treat industrial effluents from petrochemical, dairy, meat processing, abattoir, and pulp and paper factory production. Brewery, winery, tannery and olive mills wastewaters have been recently added to CW applications. CWs can be applied to several and different kinds of industrial wastewaters, including acid mine wastewater with low organic matter content and landfill leachate. Vymazal [9] reported the use of CWs for the treatment of industrial wastewaters with influent concentrations up to 10,000–24,000 mg of chemical oxygen demand (COD)/L and up to 496 mg NH_4^+/L. However, there are no general rules for selecting the most suitable type of CW for a certain industrial wastewater or even urban wastewater. Every single case must be studied according to several conditions: type of wastewater, land availability, amount of flow and pollutant load, outlet discharge limits, etc. [8].

Industrial wastewaters differ substantially in composition from municipal sewage, as well as among themselves. Industrial wastewaters can present very high concentrations of organics, total suspended solids (TSS), ammonia and other pollutants; therefore the use of CWs almost always requires some kind of pretreatment. The BOD/COD ratio is a parameter which tentatively indicates the biological degradability. If this ratio is greater than 0.5, the wastewater is easily biodegradable, such as wastewaters from dairies, breweries, the food industry, abattoirs or starch and yeast production. The BOD/COD ratio for these wastewaters usually ranges between 0.6 and 0.7 but could be as high as 0.8. On the other hand, wastewaters with a low BOD/COD ratio and, thus, low biodegradability are represented, for example, by pulp and paper wastewaters. Tentative comparison of the industrial wastewater strength with municipal sewage could be done on the basis of population equivalent (PE: 60 g BOD_5 per person per day).

3.2 Aerated Constructed Wetlands

Oxygen availability to support aerobic processes is the main limitation in HF CWs, especially when nitrification (and subsequent total nitrogen removal) is a treatment objective [10]. As a result, to increase the oxygen availability, CWs have evolved into more effective treatment systems by installing an aeration system capable of transferring sufficient oxygen to perform aerobic processes. Design variants now span from completely passive systems (HF), to moderately engineered systems (unsaturated VF systems with pulse loading) up to highly engineered or intensified systems, with increased pumping, water level fluctuation, or forced aeration [11].

As a result, most of the treatment wetland design and operational modifications developed in the last decade aim at improving subsurface oxygen availability. The simplest (most passive) modification is the construction of shallow HSF flow beds, highlighted by Garcia et al. [12]. Their findings suggest that by limiting the depth of the HF bed, all of the wastewater is forced through the root zone. Their results show improved treatment performance for COD, BOD_5, and NH_4-N in shallow beds (27 cm water depth) compared to deeper ones (50 cm water depth). However, recent studies suggest that this positive effect of shallow beds is limited to low surface loading rates [13, 14].

Recirculation of treated effluent has also been shown to improve removal of ammonium nitrogen and organic matter [15–19]. Operational adaptations to improve subsurface oxygen availability include water level fluctuations such as batch loading [20–22], "fill-and-drain", "reciprocating", or "tidal flow" [23–28]. A step forward is the use of active aeration (e.g., a network of air distribution pipes installed at the bottom of the bed connected to a blower pump to supply atmospheric air) which has also been applied to HF to constructed wetland beds [29–32] and saturated VF systems [33, 34], often showing a more than ten-fold increase of removal rates compared to passive systems. Most of the reports on intensified treatment wetland designs come from private engineering companies which hold patents. However, the potential use of intensified treatment wetland is widely recognized, and design guidance and parameters have yet to be determined [35].

As indicated, VF CWs have predominant aerobic conditions, while HF CWs mainly presented anaerobic conditions. Combining both types of CW in hybrid systems could achieve complete nitrogen removal, so in more recent years, interest in the study of multi-step and hybrid systems has increased [9, 36, 37]. The most commonly used hybrid system is the two-step VF-HF CW, which has been used for treatment of both sewage and industrial wastewaters [9, 38, 39]. In general, all types of hybrid CWs are comparable with single VF CWs in terms of NH_3-N removal rates whilst they are more efficient in TN removal than single HF or VF CWs [9]. However, even in hybrid VF+HF systems, the TN removal remains low [9, 40, 41]. The effectiveness of alternating aerobic and anaerobic conditions in VF-HF hybrid systems was evaluated by Gaboutloeloe et al. [40], who reported that the most limiting factor of these systems was nitrate accumulation, mainly caused by the depletion of carbon during the aerobic phase. Tanner et al. [41] pointed out that the endogenous organic carbon supply from plant biomass decay and root-zone exudation has often been found to be insufficient to achieve full denitrification in VF+HF hybrid systems. In order to solve this handicap and improve TN removal, several authors studied the effect of step-feeding in tidal and saturated VF CWs [42–45]. Tanner et al. [41] proved the use of carbonaceous bioreactors, which incorporate a slow-release source of organic C (e.g., wood chips) aiming to increase denitrification. Recirculation has been employed in various configurations [46–50] in order to increase simultaneous nitrification and denitrification processes in either a single CW unit or in the two-step HF+VF system. Artificial aeration in hydraulic saturated units, attaining to only part of the system or timed, has a high potential as an alternative to enhance TN removal [32, 51–53].

3.2.1 Oxygen Transfer at the Water–Biofilm Interface

Early HF wetland designs were based on the Root Zone Method (RZM) [54]. Plant-mediated oxygen transfer was thought to be a key mechanism in RZM designs, but actual oxygen transfer rates generally did not meet these design expectations [55] and the systems often clogged. This led to the development of VF wetlands in the late 1980s. However, if VF wetlands are hydraulically or organically overloaded, ponding of wastewater occurs. This effectively cuts off air circulation and promotes clogging, which dramatically reduces oxygen transfer [56].

Mechanisms for oxygen transfer in treatment wetlands include atmospheric diffusion, plant-mediated oxygen transfer, and oxygen transfer at the water–biofilm interface [10]. Research in the recent years identified several design and operation factors, which improve oxygen transfer at the water–biofilm interface, such as artificial aeration [51, 52, 57, 58] and fill-and-drain operations [3, 59]. Since the rate of air circulation (and thus oxygen transfer) is related to the frequency of water level fluctuation in filling and draining systems, internal recycling to rapidly fill and drain multiple wetland compartments is often employed.

From a pilot-scale research facility in Langenreichenbach, Germany, Nivala et al. [10] estimated oxygen consumption rates (OCR) for the main CW designs. Measured OCRs ($g/m^2/d$) were in the range of 0.5–13 for HF CW, 8–59 for VF CWs and 11–88 for intensified CW systems. Similar or even higher OCRs were previously reported in literature. However, as pointed out by Nivala et al. [10], those rates may not necessarily be sustainable over the long term operation of the system. Intensifying oxygen input in the CWs through the use of artificial aeration combine the advantage of maintaining low energy consumption and clogging prevention [31]. Artificial aeration strategies can vary extensively from partial to total aeration in relation to time and space, and from low to high intensity. Although some small and large field applications of aerated CW have been reported, properly described experiences of artificial aerated CWs are mainly limited to a few laboratory and pilot scale systems [31, 32, 51, 52, 57].

Aerated HCW varied from 0.3 m to 1 m in depth [32, 51, 52]. Probably, an efficient aeration process requires a higher depth in order to reach a high oxygen transfer rate (OTR) enhanced by sufficient long contact time between the supplied air and WW. Automated aeration devices were used in some cases, setting dissolved oxygen (DO) concentration set point to activate air pumps in the range of 0.2–0.6 mg/L [51]. In other cases, continuous aeration was provided, over the overall wetland bed surface [10, 52] or only near the inlet zone [32]. However, intermittent aeration or a spatial segregation of aerated and non-aerated zones has been considered possible in order to reach simultaneous nitrification and denitrification [32, 43, 53, 60, 61]. In efficient aerated HF CW, nitrification occurs when the aeration system is turned on, while denitrification requires anoxic conditions, which could be obtained by ceasing aeration. Aeration intensities were reported for VF CW by Pan et al. [62] and Maltais-Laundry et al. [32] ranging from 0.12 to 0.76 $m^3/m^2/h$. These authors found that oxygen utilization efficiency decreased when the aeration intensity increased.

3.2.2 Benefits of Artificial Aeration in Constructed Wetlands

Important factors affecting the treatment performance include the flow type, substrate characteristics, plant species, hydraulic loading rate (HLR) and temperature. HLR, related to the space available for the water to flow through the CW, is a principal parameter for the design and operation of CW. Sakadevan and Bavor [63] reported that the removal of pollutants in a CW was improved by decreasing HLR when the applied hydraulic retention time (HRT) ranged from 4 to 15 days. A lower HLR implies more contact time and more treatment stability, however, it occupies a larger land area [5].

Physical processes such us sedimentation and decantation, important in particulate organic matter removal, are mostly unaffected by winter conditions. However, biological processes are temperature dependent, and winter removal performances of HF CWs for nitrogen and soluble organic matter, both highly driven by biological activity, may be reduced [59, 64].

Besides lower winter temperature, low oxygen availability, which is already a common limiting factor in HF CWs during the growing season, may be even more so in winter. Oxygen solubility is higher in colder water [65], but gas exchange in HF CWs may be reduced by the additional insulation layer and the fact that plants are dormant. Low oxygen content results in low aerobic organic matter decomposition [28, 66–68]. This leads to fermentation processes [69, 70] that can represent, in certain overload cases, the main way of organic matter decomposition [71]. Moreover, the nitrification step represents the main limiting factor for N removal in HF CWs because of low oxygen availability [72]. In addition, to the TN concentration, the form of N is also often a crucial factor affecting the receiving water body. For instance, besides being toxic to aquatic biota, the associated nitrogenous biochemical oxygen demand of NH_4^+-N can depress DO levels.

Although in CWs, oxygen availability may be enhanced by the presence of macrophytes through diffusion of oxygen via the aerenchym to the rhizomes [66], the exact contribution of plants remains in debate [52, 73–76]. It was reported that the contribution of plants to pollutant removal was usually less than 10% [77], although it has been found to be important for nutrient removal in low loaded systems [77, 78]. Caldheiros et al. [80] also found that there was no significant difference in pollutant removal between the planted and unplanted wetlands during a 17-month operation period. The primary role of plants is to hold the wetland components in place, preventing erosion and landscape integration. Therefore, artificial aeration appears necessary when the CW is operated under a high HLR.

3.2.3 Dissolved Oxygen Profile along CWs

DO plays an important role in the activity of microbes in wetlands. To achieve the simultaneous removal of organic matter (COD) and nutrients (N, P), the aerobic and anoxic regions in wetlands need optimization depending on wastewater characteristics and operational manipulation.

Dong et al. [58] compared different aeration strategies in three VF CWs: non-aeration (NA), continuous-aeration (CA) and intermittent-aeration (IA), to treat heavily polluted river water. The VF CWs were continuously fed from a feed tank using a metering pump. Although the VF CWs have higher oxygen mass transfer efficiency than the HF ones, the DO concentrations (averaged over three tested HLRs) in the 5–40 cm region above the reactor bottom were below 1 mg/L in NA, which could inhibit the nitrification process.

For the CA, DO concentrations ranged from 1.3 to 2.2 mg/L (in the 5–20 cm profile) and from 3.8 to 4.4 mg/L (in the 40–60 cm profile). However, for the IA, DO concentrations varied from 0.8 to 1.1 mg/L and from 2.5 to 2.8 mg/L in the mentioned DO profiles.

It was reported that no obvious nitrification was observed when the DO concentration was lower than 0.5 mg/L [81]. According to the DO values, artificial aeration significantly improved the oxygen availability in the VF CWs. Although all DO concentrations in IA and CA appeared to exceed that required for anoxic condition (i.e., 0.2–0.5 mg/L), anoxic regions could still exist in the aerated VF CWs due to the spatial stratification of biofilms in both IA and CA operation modes, and particularly in the IA operation mode, which would facilitate denitrification.

3.2.4 TSS Removal

Several authors found that supplemental aeration of CWs had a positive effect on TSS reduction [82, 83]. Ouellet-Plamondon et al. [31] concluded that artificial aeration may have reduced matter accumulation by increasing degradation kinetics and prevented clogging.

3.2.5 COD Removal

COD removal is related to HLR. The increase of HLR makes the removal efficiencies of COD decrease. Increasing HLR would reduce the contact time between wastewater and microbes, enhance the detachment of microbes off substrate surfaces, and decrease the oxygen availability [84]. Although organic matter can be degraded both aerobically and anaerobically by heterotrophic bacteria in the wetlands depending on local DO concentrations, aerobic degradation is usually more important [85]. Dong et al. [58] found that COD removal efficiency was positively correlated with the aeration condition: CA > IA > NA, continuous-aeration, intermittent-aeration and non-aeration conditions, respectively (see Section 3.3.3).

As reported by Ouellet-Plamondon et al. [31], during summer, there was a slight improvement in COD removal in planted mesocosms compared to unplanted ($p < 0.01$), but no effect of artificial aeration, regardless of the presence of plants. In winter, the expected reduction in COD removal in non-aerated mesocosms was totally compensated with a significant improvement in aerated mesocosms, both for planted and unplanted units. The added oxygen in winter probably counterbalanced the reduction of removal kinetics due to temperature and plants dormancy [31].

When oxidation decreases, the amount of residual inert organic matter accumulated increases and aggregates in filtration matrix changing the hydraulic conditions by reducing HRT [3, 84] and biological properties [87]. Increasing oxygen availability with artificial aeration could enhance mineralization and reduce hydraulic clogging [88]. Sulphate reduction, a typical diagnostic of poor oxygen conditions in CWs [89], could also be inhibited by artificial aeration. Thus, for organic matter removal, their results suggest that artificial aeration in HF CWs could be beneficial in winter, when plants are dormant.

3.2.6 Nitrogen Removal

Nitrogen removal in CWs occurs through adsorption, assimilation into biomass, ammonia volatilization and coupled nitrification/denitrification, of which the nitrification/denitrification process is the most important [90, 91].

Since nitrification and denitrification are two operationally separate processes (either temporally or spatially), which respectively require aerobic and anoxic conditions, the rate of nitrification significantly impacts the removal of TN. The removal efficiency of TN significantly dropped with an increase of HLR. Artificial aeration significantly improved the oxygen availability and thus enhanced the removal of NH_4-N in the VF CWs. Intermittent aeration was optimal for TN removal, which facilitated denitrification due to both spatial and temporal formations of anoxic zones in the VF CWs. Although continuous aeration achieved the highest nitrification rate, the denitrification process was notably suppressed due to an excessive oxygen supply that artificial aeration significantly enhanced NH_4-N removal in VF CWs [53, 58].

Besides all this, average TKN removal in winter was lower than in summer, most likely because of the lower winter temperature, which was well under optimal temperature for nitrifying activity [92]. In winter, artificial aeration improved TKN removal for all mesocosms ($p < 0.01$), with a more pronounced improvement for unplanted units [31].

3.3 HIGHWET Project

The HIGHWET project was addressed to improve the capacity and effectiveness of CWs as high-rate and sustainable wastewater treatment system. HIGHWET aimed to perform and validate new approaches based on the combination of the hydrolytic up-flow sludge bed (HUSB) anaerobic digester and CWs with forced aeration for decreasing the required surface of conventional HF CWs and improving the final effluent quality. For this purpose, two demonstration plants were designed and constructed in Spain and Denmark. The first configuration (A Coruña, NW of Spain) consisted of a HUSB and HF CWs for raw municipal wastewater treatment [53], while second configuration (at KT Food, nearby Aarhus, Denmark) consisted of a combination of a HUSB and hybrid (FV-HF) CWs for treatment of high load organic industrial wastewater. The effect of effluent recirculation, aeration regime and different phosphorus adsorbent materials was planned to be checked in both plants. The authors report in this chapter the results obtained in the KT Food HIGHWET plant.

3.3.1 KT Food Pilot Plant

KT Food is a food producing company located at Randersvej 147, in the town of Purhus (56º 33' 38.58" N 9º 51' 26.08" E) in Denmark. Additional to the production of food, the site also generates water from a small dairy farm and domestic activities. To meet the discharge standards demanded by the environmental authorities, a wastewater treatment system was built. The plant was constructed in August 2014 as a research plant funded by the European Union under the FP7 grant agreement N° 605445. After technical, environmental considerations and to meet the discharge demands, it was decided to design and construct the treatment plant using a combination of an anaerobic digester as primary treatment, followed by two parallel treatment trains (aerated line and non-aerated line) of constructed wetlands, several wells to allow controlled recirculation of treated waters and additional wells to host reactive media to remove P before discharge The conceptual design of the system installed is shown in Figure 3.1.

The wastewater is collected from the house and the two industrial plants, homogenized in a well where fat and grease is removed. Thereafter, wastewater is pumped to the treatment plant where flow is measured using an ultrasound digital flow meter. Once the research project is finished, all the wastewater produced at the site is being treated by the plant.

After the first well, water is transported to a second well where pollutant concentration can be increased by adding a prepared feedstock solution in order to reach the desired concentrations for the research project. The primary treatment consisted of a Hydrolytic Upflow Sludge Blanket (HUSB) digester. After the HUSB, water flows to a pumping well that is fitted with two pumps, where direction and flow volume can be selected to any of the two treatment trains. Both treatment trains consist of a VF CW followed by HF beds and phosphorous removal wells. The surface of the VF beds is 16 m^2, while both of the HF beds are 3 m^2. In the eastern train, the VF bed is fitted with forced aeration. while the bed of the western train is passively aerated. Both of the HF beds are fitted with forced aeration. The aeration systems installed to supply air to the beds use individual compressors that provide atmospheric air to increase the oxygen availability, and improve the aerobic processes of pollutant degradation. Additionally, the aeration time and cycles to each one of the beds can be controlled with automatic timers according to the operation planned.

3.3.2 Research Operational Plan of KT Food Treatment Plant

During the research phase, the WWTP operated with different loading schemes where pollutant loadings, aeration cycles and recirculation were modified to obtain the largest amount of data possible and to determine the treatment capacity. Sampling strategy was to take grab samples from nine different points along the treatment train (Figure 3.1). Five sampling campaigns were performed

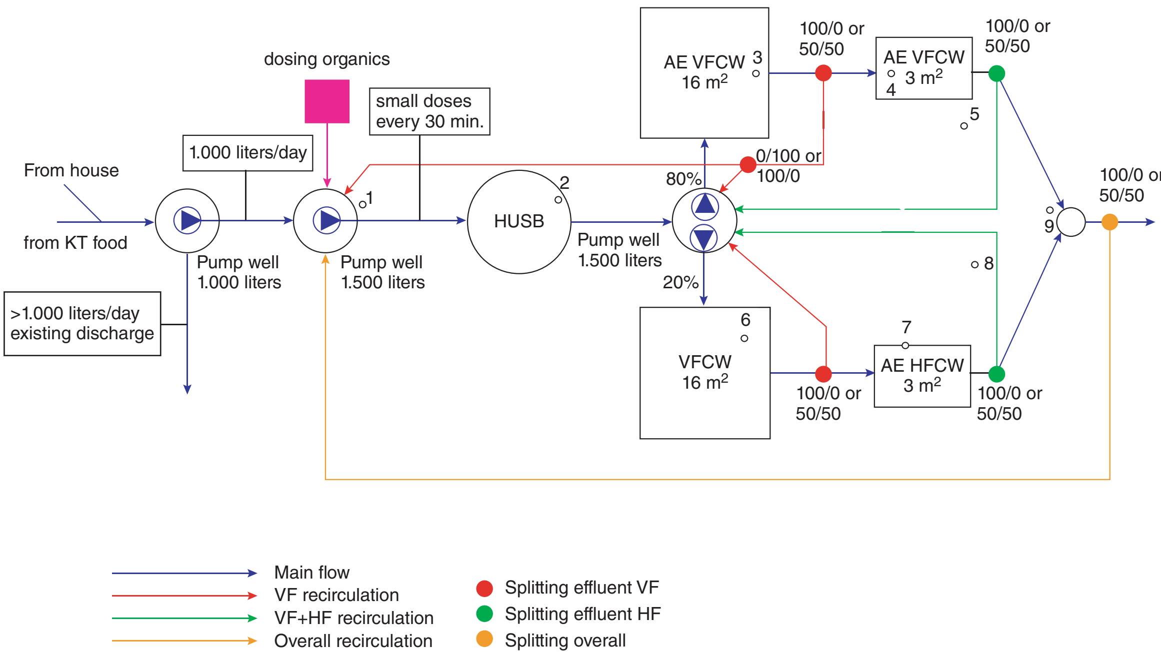


Figure 3.1 Conceptual design of the system installed at KT Food. Empty circle and numbers indicate the sampling points along the treatment trains 1) inlet; 2) after the HUSB; 3) after the aerated VF; 4) after the aerated HF bed; 5) after the P removal filtered filled with Tobermorite; 6) after the non-aerated bed; 7) after the aerated bed; 8) after the P removal filter filled with Polonite; 9) final effluent. Treatment trains: aerated line (1–2–3–4–5) and non-aerated line (1–2–6–7–8).

Table 3.1 Planned exploitation parameters for each of the sampling campaigns.

Operation parameter[a]	Campaign 1	Campaign 2	Campaign 3	Campaign 4	Campaign 5
Month	January	March	May	July	September
AE VF CW – AE HF CW (L/d)[b]	800	800	800	800	1440
VF CW – AE HF CW (L/d)[c]	200	200	200	200	360
Recirculation	No	No	No	No	80%
VF CW aeration time (h on/h off)	24/0	24/0	4/4	6/2	24/0
HF CW aeration (h on/h off)	24/0	24/0	24/0	24/0	24/0

[a] Planned influent concentration was 5000 mg COD/L, 500 mg TN/L and 30 mg TP/L along the whole study.
[b] AE VF CW – AE HF CW = Aerated Vertical Flow Constructed Wetland – Aerated Horizontal Flow Constructed Wetland.
[c] VF CW = Vertical Flow Constructed Wetland.

for different aeration schemes and effluent recirculation (Table 3.1). Analyses were carried out as described in Standard Methods [91].

Any change of operational parameters implies the need of acclimation time so that the processes can become stable and performance is optimized. Therefore a period of three to four weeks acclimation time was allowed between the measuring campaigns.

After the first samples were collected during plant start-up (data not shown), it was evident that the wastewater produced at the site did not reach the aimed high concentration to achieve the organic or nutrient overloading stated in the exploitation plan. Therefore, it was necessary to install a system that could supply a prepared solution to reach the planned pollutant and hydraulic loadings. The system was built using a 1 m^3 tank, and a time controlled dosing pump that fed the solution to the well located before the HUSB. The loading solution was prepared using a blending of fresh pig manure, molasses, starch, urea and fertilizer. The volumes of each component were calculated to reach the planned loading and were monitored regularly to maintain a constant loading. The flow was controlled and always was close to the desired overall flow of 1,000 L/d.

After the initial adaptation period, plant equipment including aeration pumps and a dosing system functioned without any problems, in spite of the low temperatures and the snow that covered the system, which is to be expected for the winter period in Denmark (Figure 3.2e). As it can be seen in Figure 3.2, no plant development was present during Campaigns 1 and 2, but plant development started in April, before Campaign 3 carried out in May.

According to the exploitation plan, the last campaign included increasing the flow by recirculating 80% of the treated water that went through the aerated VF bed. That means that during the campaign, the overall influent flow to the beds was of 1,800 L/d. The flow to the forced aerated bed was 1,440 L/d and to the passively aerated bed 360 L/d, while the hydraulic loading rates were 9 cm/d and 2.25 cm/d, respectively.

Figure 3.2 Satellite image showing the location of the treatment plant (a); a general view of the plant once it was established (b); details of HF bed (c); detail of P removal unit (d); state of the system during Campaign 1 at winter 2015 (e); state of the non-aerated VF bed during Campaign 2 (f); the two VF beds during Campaign 3 (aerated VF bed: g; passively aerated VF bed: h).

3.3.2.1 Campaign 1

Campaign 1 was carried out during winter 2015. Averages and the standard deviation of the evaluated parameters are presented in the following tables (Tables 3.2–3.4). Even though environmental temperature was below or close to 0°C, the wastewater temperature was always above freezing in the beds. Temperature was uniform along the components of the treatment. Electrical Conductivity (EC) decreased along the treatment. pH was in the range of 6.7–10.6 in the different beds, being the highest after the Polonite well. As expected, DO was low at the inlet and after the HUSB. As water went through the system, the DO increased to reach oxygen saturation.

Table 3.3 presents the results of the average concentrations of TSS, COD and BOD_5 during the campaign. TSS concentration varied between 96 mg/L at the inlet to 3.9 at the effluent. The overall

Table 3.2 In-situ monitored parameters during Campaign 1.

	Temperature (°C)		EC (μS/cm)		pH		DO (mg/L)	
Sampling place	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	7.9	2.0	1,775	285	6.9	0.9	3.4	3.3
2: After HUSB	7.8	1.0	1,325	445	6.7	1.3	0.4	0.2
3: After aerated VF bed	7.4	1.8	817	208	8.0	0.2	10.2	1.8
4: After HF bed	6.8	1.7	697	203	8.3	0.3	12.1	0.8
5: After Tobermorite	6.7	1.5	721	182	9.3	0.1	9.0	1.9
6: After non-aerated VF bed	6.8	1.9	733	233	8.1	0.2	8.6	2.5
7: After HF bed	6.3	1.9	566	180	8.2	0.2	12.4	1.0
8: After Polonite	6.2	1.9	504	156	10.6	0.5	10.3	0.8
9: Effluent	6.5	2.1	597	188	9.8	0.5	9.0	2.0

Table 3.3 Average TSS, COD and BOD_5 in the system during Campaign 1.

	TSS (mg/L)		COD (mg/L)		BOD_5 (mg/L)	
Sampling place	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	96	49.17	5538	381	2567	603
2: After HUSB	40	12.02	2541	459	1317	580
3: After aerated VF Bed	12	7.3	50	36	4.0	1,0
4: After HF bed	4.8	1.3	70	21	5.0	6.1
5: After Tobermorite	5,8	1.6	39	21	18.0	14.7
6: After non-aerated VF bed	8	2.2	119	33	12.7	9.6
7: After HF bed	5	1.4	52	29	3.3	3.2
8: After Polonite	8	3.2	63	35	3.3	4,0
9: Effluent	3.9	4.6	51	16	4.7	2.5

Table 3.4 Average nutrient concentrations and performance along the system during Campaign 1.

	NH_4-N (mg/L)		NO_3-N (mg/L)		TN (mg/L)		TP-P (mg/L)	
Sampling place	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	116	69	49	14	489	71	32	17.6
2: After HUSB	87	82	12	15	232	7	22	8.3
3: After aerated VF Bed	0	0	8.8	7.5	54	2	3	0.7
4: After HF bed	0	0	8.5	6.5	58	3	2	0.4
5: After Tobermorite	1	2	8.5	7.1	50	2	2	0.4
6: After non-aerated VF bed	1	1	6.7	5.3	121	2	1	0.2
7: After HF bed	0	0	8.1	4.9	83	5	0.8	0.4
8: After Polonite	0	1	4.2	2.3	66	2	0.4	0.2
9: Effluent	0	1	2.8	2.8	40	0	1	0.3

removal of TSS was 96% while the reduction in the HUSB was around 60%. There is further reduction along the system and the final concentration is sufficient to meet any discharge standard. A high reduction of COD occurred in the HUSB where 50% of the COD was removed. Further COD removal happened in the aerated bed reaching 98% removal. The removal between the HUSB and the non-aerated bed was also high, reaching 95%. After the two VF beds, there were low removal but it can be explained by the low COD concentrations after the VF beds. Average BOD_5 concentration during the campaign at the influent was around 2,600 mg/L and around 5 mg/L at the effluent, with an overall removal of 99%. Between the influent and the HUSB, the removal of BOD_5 reached 49%. After the HUSB, the removal of BOD_5 reached 99%, both in the aerated VF bed and in the non-aerated bed.

Conversion of nitrogen compounds and total phosphorus is given in Table 3.4. Nitrification in the system was effective and the overall nitrification was close to 100%. The nitrification process occurred mainly while the water was in the VF beds. Simultaneous to nitrification, denitrification was also taking place along the treatment and the overall denitrification rate was 94%. Denitrification occurred in the HUSB where 75% of the NO_3-N was removed. P removal in the system occurred in all the structures, reaching up to 97%. The two reactive materials tested showed that they can produce effluents with concentrations below 1 mg/L.

3.3.2.2 Campaign 2

The results obtained for Campaign 2 are presented in Tables 3.5–3.7. Table 3.5 shows the average temperature along the structures which are affected by the external temperature, ranging from around 12°C to 9°C. EC was higher at the influent and decreased along the treatment. pH was around 8 but increased above 11 after the Polonite tank. DO concentration was low at the influent but increased along the treatment to reach nearly DO saturation concentrations after the first VF beds in both treatment trains.

In spite of TSS concentration in the influent was higher than the previous campaign, TSS concentration after the HUSB was already 51% lower (Table 3.6). Further removal occurred along the treatment reaching an overall TSS removal of 97%. COD was around 6,000 mg/L with an overall removal of 99% at the effluent. The HUSB removed 71% and the VF beds were able to remove the rest of the COD.

Table 3.5 In-situ measured parameters during Campaign 2.

	Temperature (°C)		EC (µS/cm)		pH		DO (mg/L)	
Sampling place	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	11.8	0.9	1,773	549	7.8	0.5	1.6	2.1
2: After HUSB	10.2	0.9	1,503	212	8.1	0.5	0.8	0.5
3: After aerated VF Bed	10.4	0.2	1,214	266	8.6	0.1	12.0	0.2
4: After HF bed	9.6	0.6	1,222	266	8.6	0.1	9.7	0.4
5: After Tobermorite	9.3	0.5	982	203	9.3	0.1	9.7	0.4
6: After non-aerated VF bed	9.8	0.5	1,490	413	8.2	0.1	9.3	0.6
7: After HF bed	9.2	1.0	962	228	8.6	0.2	12.1	0.3
8: After Polonite	8.8	0.6	709	87	11.2	0.1	10.7	0.4
9: Effluent	9.4	0.5	829	320	9.8	0.5	10.0	1.0

Table 3.6 Average TSS, COD and BOD_5 in the system during Campaign 2.

	TSS (mg/L)		COD (mg/L)		BOD_5 (mg/L)	
Sampling place	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	235	56	6108	1547	3500	1061
2: After HUSB	115	20	1748	161	1625	530
3: After aerated VF Bed	19	8,0	57	8	8,5	3,5
4: After HF bed	6,7	3,1	49	5	1,5	0,7
5: After Tobermorite	8,4	0,4	45	5	1,0	0,0
6: After non-aerated VF bed	11	7,5	67	20	3,0	1,4
7: After HF bed	6	1,8	37	1	2,5	0,7
8: After Polonite	10	3,2	28	2	1,0	0,0
9: Effluent	8,2	1,0	38	3	1,0	0,0

Similarly, BOD_5 at the influent was on average 3,500 mg/L with a removal of 54% in the HUSB. The VF beds removed on average more than 99% of the remaining BOD_5 leaving very little BOD to be removed in the following structures.

Regarding nutrient removal (Table 3.7), different behavior took place compared to the previous campaign. Dynamics of the removal along the beds were different than in previous campaigns, reaching an overall removal of 94%. NH_4-N average concentration in the inlet was around 100 mg/L, with only 10% removal in the HUSB. In the aerated VF the removal was close to 60%. The NH_4-N removal in the non-aerated bed was less effective and only 46% was removed. The rest of the system continued to remove NH_4-N to reach a final concentration of 6 mg/L. Nitrate in the system was removed in all the structures, especially in the HUSB, where all the NO_3-N was removed. Along the

Table 3.7 Average nutrient concentrations (mg/L) and performance along the system during Campaign 2.

	NH_4-N		NO_3-N		TN		TP-P	
Sampling place	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	101	46	26	5.3	152	61	26	12.9
2: After HUSB	89	4	1.1	0.2	89	34	16	3.3
3: After aerated VF Bed	34	9	23	8.9	58	11	5	0.3
4: After HF bed	25	4	24	3.8	61	9	4	0.3
5: After Tobermorite	9	6	21	1	38	3	2	0.2
6: After non-aerated VF bed	48	32	33	13.4	89	13	3	0.9
7: After HF bed	7	6	24	5.4	30	6	1	0.1
8: After Polonite	0	0	16	1.6	19	1	0.4	0.1
9: Effluent	6	5	18	0.8	28	2	2	0.7

bed there was an increase of NO_3-N as a result of nitrification in the structures. Both of the tested media presented good P removal capacity, reaching 92% through the overall system.

3.3.2.3 Campaign 3

After Campaign 2, aeration time for the aerated VF bed was set to intermittent aeration (4 hours on, 4 hours off). The results obtained Campaign 3 are presented in Tables 3.8–3.10. The third campaign took place in May when temperatures began to increase and weather was milder. The plants in all the beds began to grow due to the noticeable effect of the season being more effective in the aerated beds because of the higher water flow that allowed better nutrient availability. Even though weather was milder, it was not reflected in the water temperature. This can be explained by the low temperatures

Table 3.8 In-situ monitored parameters during Campaign 3.

	Temperature (°C)		pH		EC (µS/cm)		DO (mg/L)	
Sampling points	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	12.1	0.6	7.6	0.6	1883	746	0.7	0.4
2: After HUSB	10.3	0.5	7.7	0.3	1539	289	0.7	0.4
3: After aerated VF Bed	10.5	0.2	8.0	0.1	1394	140	6.3	0.9
4: After HF bed	9.6	0.7	8.7	0.0	1352	64	11.9	0.1
5: After Tobermorite	9.6	0.4	9.3	0.0	1126	65	9.5	0.2
6: After non-aerated VF bed	9.9	0.6	8.2	0.1	1671	244	9.1	0.6
7: After HF bed	9.1	1.2	8.7	0.0	1063	130	12.0	0.3
8: After Polonite	8.4	0.5	11.3	0.0	763	50	10.6	0.3
9: Effluent	9.3	0.6	9.6	0.3	981	124	9.6	0.8

Table 3.9 Average TSS, COD and BOD_5 concentrations in the system during Campaign 3.

Sampling point	TSS (mg/L)		COD (mg/L)		BOD_5 (mg/L)	
	Aver	Stdv	Aver	Stdv	Aver	Stdv
1: Inlet	117	14	5,268	917	4,167	4,404
2: After HUSB	25.2	6.4	2,055	1516	1,383	693
3: After aerated VF bed	22.6	26.8	211	37.5	44	25
4: After HF bed	33.6	21.7	156	13.5	6	2
5: After Tobermorite	33.8	31.7	156	6.8	6	2
6: After non-aerated VF bed	86.5	23.0	174	60	3	2
7: After HF bed	101.7	39.3	82	6.1	2	0
8: After Polonite	102.7	36.9	61	2.9	2	2
9: Effluent	47.2	31.0	130	24.0	3	3

Table 3.10 Average nutrient concentrations and performance along the system during Campaign 3.

Sampling Point	NH_4-N (mg/L)		NO_3-N (mg/L)		TN (mg/L)		TP (mg/L)	
	Aver	Stdv	Aver	Stdv	Aver	Stdv	Aver	Stdv
1: Inlet	250	162.2	113.5	16.5	382	404	25.1	18.5
2: After HUSB	197	67.4	3.6	4.6	201	419	18.2	12.2
3: After aerated VF Bed	5.2	2.3	1.2	0.1	11	12	5.2	0.4
4: After HF bed	0.12	0.021	5.0	0.6	9	4	4.0	0.3
5: After Tobermorite	0.14	0.019	4.5	0.6	11	19	2.2	0.1
6: After non-aerated VF bed	0.10	0.010	34	7.6	42	6	2.9	0.2
7: After HF bed	BDL	BDL	36	5.7	40	12	1.2	0.1
8: After Polonite	0.7	0.0	40	6.5	45	21	0.5	0.1
9: Effluent	0.3	0.0	26	6.6	27	11	1.8	0.1

reached at night. DO concentrations were as expected, with anaerobic conditions in the inlet and at the outlet of the HUSB. Except for the aerated VF bed, DO concentrations were close to saturation. The lower DO in the aerated VF bed effluent can be explained by the fact that during this campaign, aeration was carried out in cycles, with 4 hours of aeration and 4 hours with no aeration. Even though DO was lower, it was still above 60% saturation.

TSS in the raw water was within the expected limits of raw wastewater (Table 3.9). At the beginning of this campaign, a problem with the HUSB occurred, due to the presence of grease in the water surface. It was rapidly skimmed and removed from the reactor before the sampling campaign. The final effluent was about 34 mg/L. The aerated VF bed produced effluent COD concentration higher than the passively aerated bed. However, it should be considered that the aerated bed was loaded with four times the loading compared to the passively aerated bed. The targeted COD at the inlet was as planned and removal in the HUSB was effective. After the HUSB and through the two treatment trains, removal of COD was high with similar concentrations in the effluent of both VF beds. During this campaign, BOD concentration was higher than the previous campaign. This can be explained by possible changes in the food processing, as the loading solution was prepared as usual. The HUSB removed more than half of the BOD concentration. Along the system BOD was removed to low concentrations. The highest concentration of 45 mg/L was observed in the aerated VF bed.

Nitrogen species concentrations were below the targeted 500 mg/L TN (Table 3.10). This can be explained due to uncertainty about the actual concentrations of the pig manure used to prepare the solution. It can vary depending on the storage, weather conditions and the washing practices in the farm. Removal of NH_4-N was low through the HUSB. After the HUSB, removal of ammonia was effective in both trains. Results presented in Table 3.10 show that NO_3-N was denitrified in the HUSB and also in the wetlands during the shut-off of aeration periods. The passively aerated bed did not show the same effective denitrification and the effluent had a NO_3-N concentration of 40 mg/L. The same dynamics were followed by the TN.

3.3.2.4 Campaign 4

As indicated in Table 3.1, after Campaign 3, the aeration time for the aerated VF bed was set to 6 hours on, 2 hours off. Working to these conditions, the results obtained for Campaign 4 are presented in Tables 3.11–3.13. During the fourth campaign, ambient temperature increased so water temperature along the system was affected increasing to around 17°C (Table 3.11). pH behaved similarly to the previous campaigns with around 7 along the treatment and changing when water went through the P removal media. DO was low at the inlet and after the HUSB. After the aerated VF bed, DO was low but it increased as water went through the different structures of the treatment plant.

Raw water TSS was lower if compared to the previous campaign and the HUSB removed more than half of the influent TSS (Table 3.12). This suggests that skimming the grease and fat had a positive effect. The aerated bed further removed additional TSS so the concentration in the effluent was below the discharge limits. COD was around the targeted concentration and about half was removed by the

Table 3.11 *In-situ* monitored parameters during Campaign 4.

	Temperature (°C)		pH		EC (µS/cm)		DO (mg/L)	
Sampling points	**Aver**	**Stdv**	**Aver**	**Stdv**	**µS/cm**	**Stdv**	**mg/L**	**Stdv**
1: Inlet	17.0	0.3	6.8	0.7	1,690	1,196	2.2	1.9
2: After HUSB	17.3	0.4	6.2	0.5	1,059	416	1.4	1.0
3: After aerated VF Bed	17.4	0.3	8.3	0.1	1,043	82	1.5	0.4
4: After HF bed	17.3	0.4	9.2	0.0	1,039	78	8.0	0.1
5: After Tobermorite	16.8	0.2	9.7	0.0	1,070	68	5.5	1.0
6: After non-aerated VF bed	16.8	0.3	8.5	0.1	641	93	6.6	0.2
7: After HF bed	16.5	0.4	9.2	0.0	792	122	8.5	0.1
8: After Polonite	16.8	0.4	11.4	0.6	914	146	8.1	0.4
9: Effluent	17.0	0.8	10.4	0.3	991	108	7.7	0.6

Table 3.12 Average TSS, COD and BOD_5 in the system during Campaign 4.

	TSS (mg/L)		COD (mg/L)		BOD_5 (mg/L)	
Sampling point	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	117	14	4,771	1,658	2,250	0
2: After HUSB	25.2	6.4	2,516	743	1,375	106
3: After aerated VF bed	22.6	26.8	99	66.8	26	34
4: After HF bed	33.6	21.7	55	21.4	1	1
5: After Tobermorite	33.8	31.7	52	7.8	2	2
6: After non-aerated VF bed	8.5	2.3	63	26	5	1
7: After HF bed	10.7	3.3	42	10.0	6	6
8: After Polonite	10.7	3.9	37	21.2	0	0
9: Effluent	4.2	3.0	53	4.1	2	1

HUSB. The aerated bed removed around 90%. No considerable further removal was archived in this train. Through the other treatment train, the passively aerated bed performed well and was able to remove COD down to 50 mg/L. BOD followed the same pattern as COD in spite of the fact that the BOD/COD ratio was lower than in previous campaigns. The aerated bed produced an effluent, with 26 mg/L being further removed along the following structures. The treatment train with the passively aerated bed performed well reaching BOD concentrations down to 10 mg/L after the bed.

Nitrogen species in the inlet were close to the targeted concentration of 500 mg TN/L (Table 3.13). Through the HUSB there was considerable denitrification and the NO_3-N present was denitrified effectively. The HUSB did not remove NH_4-N. Nitrification seemed to be effective in the aerated bed, with inlet NH_4-N concentrations around 239 mg/L and outlet concentrations of 5 mg/L. NO_3-N was also as low as 1 mg/L suggesting that the intermittent aeration can enhance the N removal. On the

Table 3.13 Nitrogen and phosphorus concentrations in the different points during Campaign 4.

	NH_4-N (mg/L)		NO_3-N (mg/L)		TN (mg/L)		TP (mg/L)	
Sampling point	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	250	32.4	113	16.5	382	104	33.4	13.2
2: After HUSB	239	13.5	3.6	4.6	243	79	21.3	3.3
3: After aerated VF bed	5.2	2.3	1.2	0.1	7	2	5.7	1.7
4: After HF bed	0.01	0.021	5.0	0.6	6	4	5.1	0.6
5: After Tobermorite	0.01	0.019	4.5	0.6	5	2	3.9	0.6
6: After non-aerated VF bed	0.01	0.010	34	7.6	45	6	2.6	0.4
7: After HF bed	0.000	0.000	36	5.7	48	12	2.3	0.4
8: After Polonite	0.0	0.0	40	6.5	46	18	1.1	0.0
9: Effluent	0.0	0.0	26	6.6	34	21	2.3	0.7

other treatment train, wastewater was nitrified effectively, but denitrification did not occur at the same rate with intermittent aeration. No further considerable denitrification was registered in the treatment train.

3.3.2.5 Campaign 5

The fifth campaign included recirculation of the effluent of the aerated bed back to the pumping well to increase the hydraulic loading on the beds (Tables 3.14–3.16). The calculated hydraulic loading increased corresponds to around 80% more water to each one of the beds (Table 3.1). Initially and when the flow was increased, both beds presented an increase in TSS and the release of biofilm from the media was evident. A decrease in TSS concentration along time and no further biofilm was present in the effluent when the campaign started. Temperature was similar to the previous campaign

Table 3.14 In-situ monitored parameters during Campaign 5.

	Temperature (°C)		pH		EC (µS/cm)		DO (mg/L)	
Sampling point	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	16.0	0.4	6.6	1.0	2,208	1,333	1.0	0.6
2: After HUSB	15.7	0.3	6.5	0.9	1,130	228	1.7	0.3
3: After aerated VF bed	16.2	0.4	8.9	0.2	1,057	94	3.7	3.2
4: After HF bed	15.8	0.3	9.1	0.1	1,058	68	7.0	0.5
5 After Tobermorite	15.3	0.5	10.0	0.0	1,102	30	5.3	0.4
6 After non-aerated VF bed	15.1	0.4	8.3	0.1	924	118	6.2	0.6
7 After HF bed	14.9	0.6	9.1	0.4	1,163	67	8.4	0.1
8 After Polonite	14.7	0.8	11.1	0.3	1,220	44	7.8	0.4
9 Effluent	15.4	0.4	10.1	0.2	1,143	51	6.8	0.3

Table 3.15 Average TSS, COD and BOD_5 in the system during Campaign 5.

	TSS mg/L		COD mg/L		BOD_5 mg/L	
Sampling point	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	217	145	5,268	917	4,267	404
2: After HUSB	96.0	9.2	2,055	1,516	1,167	419
3: After aerated VF bed	23.3	7.6	211	37.5	22	12
4: After HF bed	7.6	4.4	156	13.5	3	1
5: After Tobermorite	9.6	5.0	156	6.8	4	1
6: After non-aerated VF bed	12.0	4.7	174	60	9	4
7: After HF bed	6.4	6.0	82	6.1	2	1
8: After Polonite	8.9	6.7	61	2.9	1	1
9: Effluent	6.9	5.6	130	24.0	3	1

Table 3.16 Average nutrient concentrations and performance along the system during Campaign 5.

	NH_4-N (mg/L)		NO_3-N (mg/L)		TN (mg/L)		TP (mg/L)	
Sampling point	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**	**Aver**	**Stdv**
1: Inlet	367	171.4	125	15.0	493	157	40	25
2: After HUSB	316	198.0	3.6	2.4	320	196	15	2.2
3: After aerated VF bed	5.9	2.1	3.2	1.2	9	1	6.3	1.2
4: After HF bed	0.00	0.00	12.9	0.5	13	0	6.2	0.5
5: After Tobermorite	3.8	1.2	26.4	9.4	30	11	5.5	0.2
6: After non-aerated VF bed	0.1	0.1	53	3.0	53	3	5.7	1.3
7: After HF bed	BDL	BDL	119	21.6	119	22	2.6	0.4
8: After Polonite	0.1	0.1	133	7.0	133	7	1.0	0.3
9: Effluent	2.9	2.1	60	17.1	63	16	1.3	0.9

because ambient temperature was mild. pH was close to neutral except when water went through the P removal material, which increased pH and in the case of Polonite up to 11. DO was low for raw wastewater and through the HUSB and relatively low after effluent of the aerated bed when measured concentration was below 4 mg/L. During this campaign 100% saturation was never achieved through the other structures.

TSS influent concentration was around 200 mg/L and the HUSB removed around 2/3 of the TSS. After the aerated VF Bed, the TSS concentration was already down to 20 mg/L. While water went through the other structures, concentration continued to drop and the final effluent was more than enough below the discharge requirements. COD influent reached the targeted concentration and the HUSB removed half of the concentration. Through the aerated bed, an additional 90% was removed and no considerable further removal occurred in the treatment train. The treatment train fitted with the non-aerated bed showed similar performance. BOD influent concentration was relatively high if compared to previous concentration, but the HUSB was able to remove around 60% of the load. The two treatment trains had no difficulty dealing with the BOD and final effluent reached concentrations close to the detection limit.

The inlet TN concentration target was reached. The HUSB did not nitrify but nitrification took place in the aerated bed. Further nitrification happened through the treatment and the wastewater was nitrified at the end of the process. Denitrification occurred in the HUSB and also in the aerated bed. After the aerated bed, no denitrification was evident. The passive aerated bed denitrified a fraction, but no further denitrification happened in this treatment train.

3.3.3 Comparison of Results

Applied SLR ($g/m^2/d$) in the aerated line of KT Food HIGHWET project were 2.5 ± 1.8, 92 ± 14, 58 ± 7, 9.1 ± 3.5, 7.8 ± 4.2 and 0.8 ± 0.1 for TSS, COD, DBO_5, TN, NH_4^+-N and TP, respectively, whilst SLRs were four times lower in the non-aerated line (i.e., 0.6 ± 0.4, 23 ± 4, 15 ± 2, 2.3 ± 0.9 and 2.0 ± 1.0 for TSS, COD, DBO_5, TN and NH_4^+-N, respectively). Thus, the non-aerated line operated at conservative design loading rates and reached satisfactory contaminant removal, usually from 90 to 99% of TSS, COD, BOD_5 and ammonia. Similar or even higher percentage removal rates were obtained in the aerated line, operated at four times higher loading rates.

TN removal reached 43 ± 7% in the HUSB digester, due to the denitrification of influent nitrate. Overall, TN removal was 85 ± 7% in the non-aerated line and 91 ± 9% in the aerated line. The aerated VF CW unit reached 80 ± 27% TN removal (91 ± 10% excluding Campaign 2), whilst the non-aerated VF CW unit reached 58 ± 36% TN removal (73 ± 17% excluding Campaign 2). TN removal was not found in the small size aerated HF units.

TP removal reached 39 ± 14% in the HUSB digester, whilst overall TP removal was 98 ± 1% in the non-aerated line and 90 ± 3% in the aerated line. Both the aerated and non-aerated VF CW units noticeable contributed to TP removal, reaching 72 ± 10% and 82 ± 13%, respectively. Additional TP removal took place in the aerated HF units, reaching 18 ± 12% and 42 ± 25% for HF1 and HF2, respectively. According to Vymazal [82], these TP removal rates obtained under average loading rates of 0.8 g TP/m^2/d may be considered very satisfactory. Finally, the P removal units with Polonite as phosphorus adsorbant material reached 56 ± 5% TP removal whilst TP removal in the unit with Tobermorite decreased from about 50% at Campaigns 2 and 3 to 11% at Campaign 5.

Therefore, the aerated line was successful in treating a four times higher loading rate and with similar or higher treatment efficiency than the non-aerated VF CW unit for organic matter and nitrogen removal and only slightly lower for phosphorus removal. The HUSB efficiently contributed to TSS, COD, BOD and nitrate removal. These high percentage removal rates were obtained at organic SLR in the range of referred studies for different kind of industrial wastewaters while ammonia and TN loading rates were higher in the aerated line of the KT Food HIGHWET plant.

Studies on industrial wastewater treatment on artificially aerated CWs are scarce. Results for coffee processing wastewater [94], dairy parlor wastewater [83], aquaculture effluent [95] and dye containing wastewater [96, 97] are summarized below.

Rossmann et al. [94] treated coffee processing wastewater (CPW) with aerated and non-aerated influent previously to pilot-scale HF CWs. The applied organic load during the experiment was 89 g COD/m^2/d, and the HRT was 12 d. Removal efficiencies of COD, BOD and TSS ranged from 87.9 to 91.5, from 84.4 to 87.7 and from 73.7 to 84.8%, respectively. Aeration of CPW in the storage tank for 2.5 days did not affect the removal efficiencies of organic matter in the CWs, which agrees with previous findings of Zhang et al. [51], due to low redox values and anoxic conditions in spite of the aeration. In this study [94], phosphorus removal (54.3–72.1%) was statistically different among treatments, with better performance for the aerated planted system, and worse for the non-aerated unplanted.

The feedlot runoff and dairy parlor wastewater in Burlington (Vermont, USA) was treated in four HF CWs (non-aerated unplanted CW1, aerated planted CW2, non-aerated planted CW3, and aerated planted CW4) of 225 m^2 each in an experiment carried out by Tunçsiper et al. [83]. HRT in CWs ranged from 3 to 16 days. Over the four years of monitoring, the CWs operated with surface loading rate of 210 g BOD_5/m^2/d and 70 g TSS/m^2/d in average. Average BOD_5 removals were 83%, 78%, 84% and 86% for CW1, CW2, CW3 and CW4, respectively. The authors of this study concluded that supplemental aeration of CWs had a positive effect on BOD_5 reduction.

Aquaculture effluent under high HLR was assessed in Jingzhou city (China) by Zhang et al. [95]. Two parallel, identical hybrid wetland systems (CW 1+2), each with down, up and HF chambers were constructed in the field. The HLR was approximately 8.0 m/day, giving a theoretical HRT of 0.96 h. For the wetland with diffused-air enhancement, there was a significant decrease in COD and NH_4^+-N concentrations after filtration. Further, the aeration significantly increased the levels of DO, ORP, nitrite, and TN, while significantly decreasing the levels of EC, COD, NH_4^+-N, and TP concentrations in the outflow compared to the non-aerated treatment. High organic loading rates of 132 and 146 g COD/m^2/d were applied for the non-aerated (stage 1) and aerated (stage 2) conditions. Concentration of COD in the effluent of aerated wetland was significantly lower than in the non-aerated wetland. TN removal was higher in the non-aerated wetland in which sedimentation of organic N was determined to be the main process of TN removal. On the other hand, TN removal was dominated by ammonium removal in the aerated stage. In the non-aerated wetland, NH_4^+-N outlet concentrations were generally higher than in the inlet, observing an opposite trend in the aerated wetland. The authors concluded that denitrification process was contained with short HRT (0.96 h) even though carbon source seemed to be enough for denitrification. Concerning P removal, higher percentage reductions were observed in the aerated wetland.

Ong et al. [97] studied the mineralization of diazo dye (Reactive Black 5, RB5) in wastewater using recirculated up-flow CW reactor in Malaysia. The HRT was 2 days. COD removal in the aerated reactor (92%) was higher than that in the non-aerated reactor (83%) whilst RB5 removal efficiency presented the opposite trend (81 and 89%, respectively).

Ong et al. [96] conducted other experiment to study the removal of azo dye Acid Orange 7 (AO7) in three parallel lab-scale CWs of 0.3 m height and 0.18 m of diameter. The CWs were planted with *Phragmites australis*, and there were aerated (A), non-aerated (B) and non-aerated unplanted (C). With an HRT of 2 days, COD removal in the aerated and non-aerated CWs was 95% and 62%, respectively (both higher than COD removal in control unplanted CW). The three CWs removed more than 94% of AO7, being slightly higher in the aerated one. The ammonia removal was significantly higher in the aerated CW (86%) than in the non-aerated CW (14%) which, additionally, performed better than the control CW (4%).

3.4 Conclusions

This study reports the effect of effluent recirculation, aeration regime and different phosphorus adsorbent materials in a system that combines a HUSB, hybrid (FV-HF) CWs and two different phosphorus adsorbent materials for treatment of industrial wastewater. Applied SLR (g/m^2/d) in the aerated line were 2.5 ± 1.8, 92 ± 14, 58 ± 7, 9.1 ± 3.5, 7.8 ± 4.2 and 0.8 ± 0.1 for TSS, COD, DBO_5, TN, NH_4^+-N and TP, respectively, whilst SLRs were four times lower in the non-aerated line (i.e. 0.6 ± 0.4, 23 ± 4, 15 ± 2, 2.3 ± 0.9 and 2.0 ± 1.0 for TSS, COD, DBO_5, TN and NH_4^+-N, respectively). The non-aerated line reached satisfactory contaminant removal, usually from 90 to 99% of TSS, COD, BOD_5 and ammonia. Similar or even higher percentage removal rates were obtained in the aerated line. TN removal reached 43 ± 7% in the HUSB digester, due to the denitrification of influent nitrate. Overall, TN removal was 85 ± 7% in the non-aerated line and 91 ± 9% in the aerated line. The aerated VF CW unit provided 80 ± 27% TN removal, whilst the non-aerated VF CW unit reached 58 ± 36% TN removal. Overall TP removal was 98 ± 1% in the non-aerated line and 90 ± 3% in the aerated line. Both the aerated and non-aerated VF CW units noticeable contributed to TP removal, reaching 72 ± 10% and 82 ± 13%, respectively. Additional TP removal was obtained in Polonite unit (56 ± 5%) at 0.2 g TP/m^2/d during the whole study whilst TP removal in Tobermorite unit at 0.87 g TP/m^2/d decreased from about 50% to 11% after 6 month of treatment. These results showed that the aerated VF CW was successful in treating a four times higher loading rate and with similar or higher treatment efficiency than the non-aerated VF CW.

Acknowledgements

This work was supported by the EU's seventh framework programme for research, technological development and demonstration under grant agreement n° 605445.

References

1 Vymazal J. Constructed wetlands for treatment of industrial wastewater: A review. Ecol Eng. 2015; 73:724–751.
2 Green M, Friedler E, Ruskol Y, Safrai I. Investigation of alternative method for nitrification in constructed wetlands Water Sci Technol. 1997; 35(5):63–70.
3 Tanner C, Sukias J, Upsdell M. Substratum Phosphorus Accumulation during Maturation of Gravel-bed constructed wetlands. Water Sci. Technol. 1999; 40:147.
4 Leibowitz SG. Isolated wetlands and their functions: an ecological perspective, Wetlands BioOne 2003; 23(3):517–531.
5 Kadlec RH, Knight RL, Vymazal J, Brix H, Cooper P, Haberl R. Constructed Wetlands for Pollution Control: Processes, Performance, Design and Operation. IWA Specialist Group on Use of Macrophytes in Water Pollution Control, Scientific and Technical Report 8; 2000.
6 Vymazal J. The use constructed wetlands with horizontal sub–surface flow for various types of wastewater. Ecol Eng. 2009; 35(1):1–17.
7 Vymazal J. Constructed wetlands for wastewater treatment: five decades of experience. Environ Sci Technol. 2011; 45(1):61–69.
8 De la Varga D, Soto M, Arias CA, Oirschot DV, Kilian R, Pascual A, Álvarez JA. Constructed Wetlands for Industrial Wastewater Treatment and Removal of Nutrients. In: Technologies for the Treatment and Recovery of Nutrients from Industrial Wastewater, 2016; pp. 202–230.
9 Vymazal J. The use of hybrid constructed wetlands for wastewater treatment with special attention to nitrogen removal: A review of a recent development. Water Res. 2013; 47:4795–4811.
10 Nivala J, Headley T, Wallace S, Bernhard K, Brix H, van Afferden M, Müller R. Comparative analysis of constructed wetlands: The design and construction of the ecotechnology research facility in Langenreichenbach, Germany. Ecol Eng. 2012; 61(B): 527–543.
11 Fonder N, Headley T. Systematic classification, nomenclature and reporting for constructed treatment wetlands. In: Vymazal J, editor. Water and Nutrient Management in Natural and Constructed Wetlands. Springer Science + Business Media B.V.: Dordrecht; 2010, pp. 191–219.
12 García J, Aguirre P, Barragán J, Mujeriego R. Effect of key design parameters on the efficiency of horizontal subsurface flow constructed wetlands Ecol Eng. 2003; 25(4):405–418.
13 De la Varga D, Ruiz I, Soto M. Winery wastewater treatment in subsurface constructed wetlands with different bed depths. Water Air Soil Pollut. 2013; 224:1485–1497.
14 De la Varga D, Díaz MA, Ruiz I, Soto M. Avoiding clogging in constructed wetlands by using anaerobic digesters as pre-treatment. Ecol Eng. 2013; 52:262–269.
15 Arias CA, Brix H. Initial experience from a compact vertical flow constructed wetland treating single household wastewater. In: Vymazal J, ed. Natural and Constructed Wetlands: Nutrients, Metals and Management. Backhuys Publishers, Leiden, The Netherlands; 2005, pp. 52–64.

16 García-Pérez A, Grant B, Harrison M. Water quality effluent from a recirculating, vertical-flow constructed wetland. Small Flows Quart. 2006; 7(4):34–38.

17 Gross A, Sklarz MY, Yakirevich A, Soares MI. Small scale recirculating vertical flow constructed wetland (RVFCW) for the treatment and reuse of wastewater Water Sci Technol. 2008; 58(2):487–494.

18 Konnerup D, Trang NTD, Brix H. Treatment of fishpond water by recirculating horizontal and vertical flow constructed wetlands in the tropics. Aquaculture 2011; 313(1–4):57–64.

19 Torrijos V, Gonzalo OG, Trueba-Santiso A, Ruiz I, Soto M. Effect of bypass and effluent recirculation on nitrogen removal in hybrid constructed wetlands for domestic and industrial wastewater treatment. Water Res. 2016; 103:92–100.

20 Stein OR, Hook PB, Beiderman JA, Allen WC, Borden DJ. Does batch operation enhance oxidation in subsurface flow constructed wetlands. Water Sci Technol. 2003; 48(5):149–156.

21 Corzo A, Pedescoll A, Álvarez E, García J. Solids accumulation and drainable porosity in experimental subsurface flow constructed wetlands with different primary treatments and operating strategies. In: Billore SK, Dass P, Vymazal P, editors. Proceedings, 11th International Conference on Wetland Systems for Water Pollution Control, Vikram University and IWA, Indore, India; 1–7 November 2008, pp. 290–295.

22 Põldvere E, Karabelnik K, Noorvee A, Maddison M, Nurk K, Zaytsev I, Mander Ü. Improving wastewater effluent filtration by changing flow regimes – Investigations in two cold climate pilot scale systems. Ecol Eng. 2009; 35(2):193–203.

23 Behrends LL. Patent: Reciprocating subsurface flow constructed wetlands for improving wastewater treatment. United States: US 5,863,433; 1999.

24 McBride GB, Tanner CC. Modelling biofilm nitrogen transformations in constructed wetland mesocosms with fluctuating water levels Ecol Eng. 2000; 14(1–2):93–106.

25 Austin DC. Patent: Tidal vertical flow wastewater treatment system and method. United States: US 6,896,805 B2; 2005.

26 Sun G, Gray KR, Biddlestone AJ, Cooper DJ. Treatment of agricultural wastewater in a combined tidal flow: downflow reed bed system. Water Sci Technol. 1999; 40(3):139–146.

27 Ronen T, Wallace SD. TAYA – Intensive wetland technology facilitates the treatment of high loads of organic pollutants and ammonia. In: Masi F, Nivala J, editors. Proceedings, 12th International Conference on Wetland Systems for Water Pollution Control, IRIDRA S.r.l. and IWA, Venice, Italy; 4–8 October 2010, pp. 872–878.

28 Wu S, Zhang D, Austin D, Dong R, Pang C. Evaluation of a lab-scale tidal flow constructed wetland performance: oxygen transfer capacity, organic matter and ammonium removal. Ecol Eng. 2011; 37(11):1789–1795.

29 Wallace SD. Patent: System for removing pollutants from water. United States: US 6,200,469 B1; 2001.

30 Higgins JP. The use of engineered wetlands to treat recalcitrant wastewaters., Southampton, United Kingdom; WIT Press. 2003: pp. 137–160.

31 Ouellet-Plamondon C, Chazarenc F, Comeau Y, Brisson J. Artificial aeration to increase pollutant removal efficiency of constructed wetlands in cold climate. Ecol Eng. 2006; 27:258–264.

32 Maltais-Landry G, Maranger R, Brisson J, Chazarenc F. Nitrogen transformations and retention in planted and artificially aerated constructed wetlands. Water Res. 2009; 43:535–545.

33 Murphy C, Cooper D. An investigation into contaminant removal in an aerated saturated vertical flow constructed wetland treating septic tank effluent. In: Vymazal J, ed. Joint Meeting of Society of Wetland Scientists, WETPOL, and Wetlands Biogeochemistry, Czech University of Life Sciences, Prague, Czech Republic; 3–8 July 2011: p. 224.
34 Wallace SD, Liner MO. Design and performance of the wetland treatment system at Buffalo Niagara International Airport. IWA Specialist Group on Use of Macrophytes in Water Pollution Control. No. 38:36–42; 2011.
35 Kadlec RH, Wallace SD. Treatment Wetlands. 2nd ed. Boca Raton, USA: CRC Press, Taylor & Francis Group; 2009.
36 Ávila C, Garfí M, García J. Three-stage hybrid constructed wetland system for wastewater treatment and reuse in warm climate regions. Ecol Eng. 2013; 61:43–49.
37 Vymazal J, Kröpfelová L. Wastewater Treatment in Constructed Wetlands with Horizontal Subsurface Flow. Dordrecht: Springer; 2008.
38 Stefanakis AI, Akratos CS, Tsihrintzis VA. Vertical flow constructed wetlands: Eco-engineering systems for wastewater and sludge treatment, 1st edn. Amsterdam, The Netherlands: Elsevier; 2014.
39 Kim SH, Cho JS, Park JH, Heo JS, Ok YS, Delaune RD, Seo DC. Long-term performance of vertical-flow and horizontal-flow constructed wetlands as affected by season, N load, and operating-stage for treating nitrogen from domestic sewage. Environ Sci Pollut. Res. 2016; 23:1108–1119.
40 Gaboutloeloe G, Chen S, Barber M, Stöckle C. Combinations of horizontal and vertical flow constructed wetlands to improve nitrogen removal. Water Air Soil Pollut. 2009; 9:279–282.
41 Tanner CC, Sukias JPS, Headley TR, Yates CR, Stott R. Constructed wetlands and denitrifying bioreactors for on-site and decentralised wastewater treatment: Comparison of five alternative configurations. Ecol Eng. 2012; 42:112–123.
42 Yang Y, Zhao YQ, Wang SP, Guo XC, Ren YX, Wang L, Wang XC. A promising approach of reject water treatment using a tidal flow constructed wetland system employing alum sludge as main substrate. Water Sci Technol. 2011; 63(10):2367–2373.
43 Hu YS, Zhao YQ, Zhao XH, Kumar JLG. Comprehensive analysis of step–feeding strategy to enhance biological nitrogen removal in alum sludge-based tidal flow constructed wetlands. Bioresour Technol. 2012; 111:27–35.
44 Fan J, Liang S, Zhang B, Zhang J. Enhanced organics and nitrogen removal in batch-operated vertical flow constructed wetlands by combination of intermittent aeration and step feeding strategy. Environ Sci Pollut Res. 2013; 20:2448–2455.
45 Wang Z, Liu C, Liao J, Liu L, Liu Y, Huang X. Nitrogen removal and N_2O emission in subsurface vertical flow constructed wetland treating swine wastewater: Effect of shunt ratio. Ecol Eng. 2014; 73:446–453.
46 Brix H, Arias CA, Johansen NH. Experiments in a two–stage constructed wetland system: nitrification capacity and effects of recycling on nitrogen removal. In: Vymazal J, ed. Wetlands – Nutrients, Metals and Mass Cycling. Leiden, The Netherlands: Backhuys Publishers; 2003, pp. 237–258.
47 Brix H, Arias CA. The use of vertical flow constructed wetlands for on-site treatment of domestic wastewater: New Danish guidelines. Ecol Eng. 2005; 25:491–500.
48 Ayaz SÇ, Aktaş Ö, Fındık N, Akça L, Kınacı C. Effect of recirculation on nitrogen removal in a hybrid constructed wetland system. Ecol Eng. 2012; 40:1–5.

49 Foladori P, Ruaben J, Ortigara ARC. Recirculation or artificial aeration in vertical flow constructed wetlands: A comparative study for treating high load wastewater. Bioresour Technol. 2013; 149:398–405.

50 Vázquez MA, De la Varga D, Plana R, Soto M. Vertical flow constructed wetland treating high strength wastewater from swine slurry composting. Ecol Eng. 2013; 50:37–43.

51 Zhang L, Zhang L, Liu Y, Shen Y, Liu H, Xiong Y. Effect of limited artificial aeration on constructed wetland treatment of domestic wastewater. Desalination. 2010; 250(3):915–920.

52 Nivala J, Wallace S, Headley T, Kassa K, Brix H, van Afferden M, Müller R. Oxygen transfer and consumption in subsurface flow treatment wetlands. Ecol Eng. 2013; 61:544–554.

53 Pascual A, De la Varga D, Arias CA, Van Oirschot D, Kilian R, Álvarez JA, Soto M. Hydrolytic anaerobic reactor and aerated constructed wetland systems for municipal wastewater treatment – HIGHWET project. Environ Technol. 2017; 38(2):209–219.

54 Kickuth R. Abwasserreinigung in Mosaikmatritzen aus aeroben und anaeroben Teilbezirken. In: Moser, F. (ed.) Grundlagen der Abwasserreinigung. MÚnchen, Wien: Verlag Oldenburg; 1981, pp. 630–665.

55 Brix H. Gas exchange through the soil–atmosphere interphase and through dead culms of Phragmites australis in a constructed reed bed receiving domestic sewage. Water Res. 1990; 24:259–266.

56 Platzer C, Mauch K. Soil Clogging in Vertical-flow Reed Beds – Mechanismns, Parameters, Consequences and …Solutions? Water Sci. Technol. 1997; 35(5):175–181.

57 Chazarenc F, Gagnon V, Comeau Y, Brisson J. Effect of plant and artificial aeration on solids accumulation and biological activities in constructed wetlands. Ecol Eng. 2009; 35(6):1005–1010.

58 Dong H, Qiang Z, Li T, Jin H, Chen W. Effect of artificial aeration on the performance of vertical-flow constructed wetland treating heavily polluted river water. J Environ Sci. 2012; 24(4):596–601.

59 Pedescoll A, Corzo A, Álvarez E, Puigagut J, García J. Contaminant removal efficiency depending on primary treatment and operational strategy in horizontal subsurface flow treatment wetlands. Ecol Eng. 2011; 37:372–380.

60 Tao W, Wang J. Effects of vegetation, limestone and aeration on nitritation, anammox and denitrification in wetland treatment systems. Ecol Eng. 2009; 35:836–842.

61 Li HB, Li YH, Sun TH, Wang X. The use of a subsurface infiltration system in treating campus sewage under variable loading rates. Ecol Eng. 2012; 38:105–109.

62 Pan JZ, Li WC, Ke F, Wang L, Li XJ. Oxygen transfer efficiency of four kinds substrates applied in artificial aeration vertical-flow wetland. In Chinese. Env Sci. 2009; 30(2):402–406.

63 Sakadevan K, Bavor HJ. Nutrient removal mechanisms in constructed wetlands and sustainable water management. Water Sci Technol. 1999; 40:121–128.

64 Kadlec RH, Reddy KR. Temperature effects in treatment wetlands. Water Environ Res. 2001; 73:543–557.

65 Stein OR, Hook PB. Temperature, plants and oxygen: how does season affect constructed wetland performance? J. Environ Sci Health A. 2005; 40(6–7):1331–1342.

66 Brix H. Constructed wetlands for municipal wastewater treatment in Europe. In: Mitsch WJ, ed. Global wetlands: old world and new. Amsterdam: Elsevier Science; 1994: 325–333.

67 Reed SC, Brown D. Subsurface flow wetlands – a performance evaluation. Water Environ Res. 1995; 67244–67248.

68 IWA (International Water Association). Constructed Wetlands For Pollution Control: Processes, Performance, Design and Operation. London: Scientific and Technical Report No 8. IWA Publishing; 2000, pp. 156.

69 Mander Ü, Kuusemets V, Lõhmus K, Mauring T, Teiter S, Augustin J. Nitrous oxide, dinitrogen and methane emission in a subsurface flow constructed wetland. Water Sci Technol. 2003; 48(5):135–142.

70 Johansson AE, Gustavsson AM, Oquist MG, Svensson BH. Methane emissions from a constructed wetland treating wastewater – seasonal and spatial distribution and dependence on edaphic factors. Water Res. 2004; 38:3960–3970.

71 Baptista JDC, Donelly T, Rayne D, Davenport RJ. Microbial mechanisms of carbon removal in subsurface flow wetlands. Water Sci Tech. 2003; 48(5):127–134.

72 Kuschk P, Wiessner A, Kappelmeyer U, Weissbrodt E. Annual cycle of nitrogen removal by a pilot-scale subsurface horizontal flow in a constructed wetland under moderate climate. Water Res. 2003; 37(1):4236–4242.

73 Bedford BL, Bouldin DR, Beliveau BD. Net oxygen and carbon dioxide balances in solutions bathing roots of wetland plants. J Ecol. 1991; 79:943–959.

74 Sorrell BK, Armstrong W. On the difficulties of measuring oxygen release by root systems of wetland plants. J Ecol. 1994; 82:177–183.

75 Armstrong W, Cousins D, Armstrong J, Turner DW, Beckett PM. Oxygen distribution in wetland plant roots and permeability barriers to gas–exchange with the rhizosphere: a microelectrode and modeling study with *Phragmites australis.* Ann Bot. 2000; 86:687–703.

76 Wie*β*ner A, Kuschk P, Kaestner M, Stottmeister U. Abilities of halophyte species to release oxygen into rhizospheres with varying redox conditions in laboratory-scale hydroponic systems. Int. J. Phytoremediation 2002; 4:1–15.

77 Olson RK. Created and Natural Wetlands for Controlling Non-Point Source Pollution. Man-Tech Environmental Technologies, Inc., USEPA, Corvallis, OR, USA; 1993.

78 Tanner C Plants as ecosystem engineers in subsurface-flow treatment wetlands. Water Sci Technol. 2001; 44(11–12):9–17.

79 Carballeira T, Ruiz I, Soto M. Effect of plants and surface loading rate on the treatment efficiency of shallow subsurface constructed wetlands. Ecol Eng. 2016; 90:203–214.

80 Calheiros CSC, Rangel AOSS, Castro PML. Constructed wetland systems vegetated with different plants applied to the treatment of tannery wastewater. Water Res. 2007; 41(8):1790–1798.

81 Vymazal J. Removal of nutrients in various types of constructed wetlands. Sci Total Environ. 2007; 380:48–65.

82 Redmond E, Just CL, Parkin GF. Nitrogen removal from wastewater by an aerated subsurface-flow constructed wetland in cold climates. Water Environ Res. 2014; 86(4):305–313.

83 Tunçsiper B, Drizo A, Twohig E. Constructed wetlands as a potential management practice for cold climate dairy effluent treatment. USA CATENA (135); 2015, pp. 184–192.

84 Toet S, Van Logtestijn RSP, Kampf R, Schreijer M, Verhoeven JTA. The effect of hydraulic retention time on the removal of pollutants from sewage treatment plant effluent in a surface-flow wetland system. Wetlands. 2005; 25(2):375–391.

85 Vymazal J. Nitrogen removal in constructed wetlands with horizontal sub-surface flow – can we determine the key process? In: Vymazal J, ed. Nutrient cycling and retention in natural and constructed wetlands. Backhuys: Leiden; 199, pp. 1–17.

86 He Q, Mankin KR. Seasonal variation in hydraulic performance of rock plant filters. Environ Technol. 2001; 22:991–999.

87 Machate T, Noll H, Behrens H, Kettrup A. Degradation of phenanthrene and hydraulic characteristics in a constructed wetland. Water Res. 1997; 31(3):554–560.

88 Carballeira T, Ruiz I, Soto M. Aerobic and anaerobic biodegradability of accumulated solids in horizontal subsurface flow constructed wetlands. Int Biodet Biodeg. 2017; 119:396–404.

89 Faulkner SP, Richardson CJ. Physical and chemical characteristics of freshwater wetland soils. In: Hammer DA, ed. Constructed Wetlands for Wastewater Treatment: Municipal, Industrial and Agricultural; 1989.

90 Nurk K, Truu J, Truu M, Mander Ü. Microbial characteristics and nitrogen transformation in planted soil filter for domestic wastewater treatment. J Environ Sci Health 2005; 40:1201–1214.

91 Saeed T, Sun G. A review on nitrogen and organics removal mechanisms in subsurface flow constructed wetlands: Dependency on environmental parameters, operating conditions and supporting media. J Environ Manage. 2012; 112:429–448.

92 Spieles DJ, Mitsch WJ. Macroinvertebrate community structure in high–and low–nutrient constructed wetlands. Wetlands BioOne 2000; 20(4):716–729.

93 APHA. Standard Methods for the Examination of Water and Wastewater. 21st edn, Washington: American Public Health Association/American Water Works Association/Water Environment Federation; 2005.

94 Rossman M, Teixeira A, Carneiro E, Fonseca F, Carraro A. Effect of influent aeration on removal of organic matter from coffee processing wastewater in constructed wetlands. J Environ Manage 2013; 128:912–919.

95 Zhang S, Li G, Chang J, Li X, Tao L. Aerated enhanced treatment of aquaculture effluent by three-stage, subsurface flow constructed wetlands under a high loading rate. Pol J Environ Stud 2015; 23(5):1821–1830.

96 Ong SA, Ho LN, Wong YS, Chen SF, Dugil DL, Samad S. Semi-batch operated constructed wetlands planted with *Phragmites australis* for treatment of dyeing wastewater. J Eng Sci Technol. 2011; 6:619:627.

97 Ong SA, Ho LN, Wong YS, Chen SF, Viswanathan M, Bahari R. Mineralization of diazo dye (Reactive Black 5) in wastewater using recirculated up-flow constructed wetland reactor. Desalination Water Treat. 2012; 46:312–320.

Annex III

Science of the Total Environment 908 (2024) 168257

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Horizontal flow aerated constructed wetlands for municipal wastewater treatment: The influence of bed depth

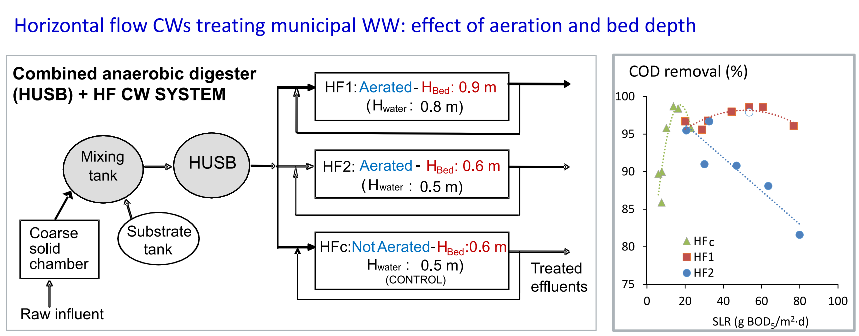
A. Pascual [a,*], J.A. Álvarez [b], D. de la Varga [c], C.A. Arias [d], D. Van Oirschot [e], R. Kilian [f], M. Soto [a]

[a] Department of Chemistry, University of A Coruña, Spain
[b] AIMEN, Pontevedra, Spain
[c] Sedaqua, Spain
[d] Department of Biology, Aarhus University, Denmark
[e] Rietland bvba, Belgium
[f] Kilian Water, Denmark

HIGHLIGHTS

- The 0.9 m deep aerated unit efficiently treated up to 80 g BOD_5/m^2d and 6.7 g TN/m^2d.
- The 0.9 m deep aerated unit at least doubled the performance of the 0.6 m deep unit.
- An aeration time of 5 h each 8 h cycle was required for advanced ammonia removal.
- Recirculation improved total nitrogen removal in aerated CWs increasing reliability.
- Methane emissions were 3 to 12 times lower for the aerated units than for the control.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Jan Vymazal

Keywords:
Constructed wetlands
Aeration
Bed depth
Nitrification-denitrification
Total nitrogen

ABSTRACT

The influence of bed depth on the performance of aerated horizontal constructed wetlands was investigated at the pilot plant scale. Two horizontal flow subsurface constructed wetlands (HF) intensified units of different bed depth (HF1: 0.90 m and HF2: 0.55 m, 0.8 m and 0.5 m water level, respectively) were fitted with forced aeration, while a third one (HFc, 0.55 m bed depth, 0.5 m water level) was used as control and not aerated. The three HF units were operated in parallel, receiving the same municipal wastewater pre-treated in a hydrolytic up-flow sludge blanket anaerobic digester. Applied surface loading rates (SLR) ranged from 20 to 80 g biochemical oxygen demand (BOD_5)/m^2·d and from 3.7 to 6.7 g total nitrogen (TN)/m^2·d, while it ranges from 6 to 23 g BOD_5/m^2·d and from 1.1 to 1.7 g TN/m^2·d in the control unit. Removal of total suspended solids (TSS) and BOD_5 was usually close to a 100 % in all units, whilst chemical oxygen demand (COD) removal was higher for the HF1 unit (97 % on average, range of 96–99 %) than for HF2 (92 %, 82–98 %) and HFc (94 %, 86–99 %). TN removal reached on average 33 % (16–43 %) in HFc, 37 % (10–76 %) in HF2 and 51 % (21–79 %) in HF1. High TN removal required a longer aeration time for nitrification and higher effluent recirculation ratio to enhance

* Corresponding author.
E-mail addresses: ana.pascual@udc.es, anapascualformoso@gmail.com (A. Pascual), jaalvarez@aimen.es (J.A. Álvarez), carlos.arias@bio.au.dk (C.A. Arias), dion@rietland.com (D. Van Oirschot), info@kilianwater.com (R. Kilian), m.soto@udc.es (M. Soto).

https://doi.org/10.1016/j.scitotenv.2023.168257
Received 14 March 2023; Received in revised form 13 October 2023; Accepted 30 October 2023
Available online 2 November 2023

denitrification. The results indicate that artificial aeration and a high bed depth allows to increase the SLR by a factor of 4 in HF1 but only by a factor of 2 in HF2.

1. Introduction

Green Infrastructure (GI), Natural Based Solutions or Ecological and Natural Infrastructure are becoming increasingly recognized as an important opportunity for addressing the complex challenges of water management while guaranteeing satisfactory treatment (Castellar et al., 2022). The GI approach refers to the natural or semi-natural systems that provide services for water resources management with equivalent or similar benefits to conventional "grey" infrastructure (Štrbac et al., 2023). The Rio + 20 Conference elevated Green Economy as one of the key features of a sustainable future. Healthy ecosystems and continuous delivery of ecosystem services are at the core of sustainable and resilient economies and growth in the transition to a Green Economy. Consequently, investments in GI have been identified as one of the main building blocks of a transition to a Green Economy (Green Infrastructure Guide, 2014). However, many GI remain relatively novel solutions, presenting important challenges and unknowns in terms of their (co) design, operation, maintenance and how to optimize their implementation.

Constructed wetlands (CWs) are GI consisting of one or more treatment cells that constitute a system designed and constructed to provide effective wastewater (WW) treatment. CWs have been in use since the 1950s and provide advanced treatment for different kinds of WW such as municipal and industrial WW, urban runoff, agricultural waste, and acid mine drainage by emulate biological, physical, and chemical processes that happen in natural wetland systems (Stefanakis, 2019; Sowińska-Świerkosz and García, 2022).

CW designs vary from passive systems to highly engineered systems that include components such as aeration, recirculation, or reciprocation (Dotro et al., 2017). The hydraulic regime in horizontal flow constructed wetlands is saturated, whereas most vertical flow constructed wetlands are generally unsaturated due to the use of pulse loading or batch feeding. Horizontal subsurface flow constructed wetlands (HF) are usually limited in oxygen availability due to permanent waterlogging of the filtration bed (Rous et al., 2019). The oxygen diffusion from the atmosphere to the waterlogged soil or sediment is very slow and slower than the use of oxygen by bacteria for oxidation of organic matter (Vymazal and Kröpfelová, 2008). Several authors have estimated root oxygen release rates from *Phragmites* spp. to be 0.02–12 $g/m^2{\cdot}d$ for horizontal flow systems, for vertical flow systems between 7.9 and 58.6 $g/m^2{\cdot}d$; and for intensified systems between 10.9 and 87.5 $g/m^2{\cdot}d$ (Zhang et al., 2014; Nivala et al., 2013). Regarding the case of HF systems, most of this oxygen is probably used to cover the respiratory demand of the root rhizome system, leaving only insignificant amounts of oxygen available for the required wastewater treatment processes (Brix, 1989).

Aerated CWs are intensified systems in which air is injected at the bottom of a saturated media filled basin. The use of an artificial aeration system dramatically increases the oxygen transfer rate compared to passive wetlands enabling improved performance for treatment reactions that require oxygen (such as organic matter removal and nitrification) or that occur more rapidly under aerobic conditions. Additionally, the aeration system can also be operated intermittently to promote nitrification/denitrification (Wallace et al., 2020). Aerated CWs have been reported to have a higher removal efficacy for nitrogen and carbon (Wallace et al., 2008) as well as pathogens (Headley et al., 2013) compared to non-aerated wetlands (Nivala et al., 2018). Comparing the treatment efficiency of several CW systems in parallel operation, these authors found that all systems achieve up to 99 % removal of suspended solids, biological oxygen demand, and ammonia, while the removal of *E. coli* ranged from 0.9 to 3.8 log units. Total nitrogen (TN) removal was partial, ranging from 17 % to 70 %, with the aerated HF and alternating CW systems achieving the highest TN removal efficiency compared to both the unsaturated and aerated vertical flow designs.

Although the knowledge published in international journals and scientific literature on the enhanced treatment performance of intensified CWs has increased in recent years, aerated CW design criteria continue to be largely a matter of patents and commercial practices (Kadlec and Wallace, 2009). There is currently no recognized design standard for aerated wetlands (Nivala et al., 2013; Vera-Puerto et al., 2022), alongside limited empirical data to support understanding of how factors, such as the configuration of aeration devices, wetland bed depth or aeration rate, impact the availability of dissolved oxygen and, ultimately, pollutant removal. Several parameters can affect the wetland aeration efficiency and, consequently, the performance of intensified treatment wetlands: gravel size and shape, plants presence, water temperature, bed and water depth, hydraulic loading rate, daily flow, surface organic loading rate, hydraulic retention time, airflow rate, total aeration time per day, atmospheric pressure, wastewater quality, fouling phenomena, maintenance of diffusers and others (Rous et al., 2019; Vera-Puerto et al., 2022). The bed depth is an important factor because it determines, among other aspects, the power demand of the air blower and the residence time of the air bubbles in the system. As pointed out by Rous et al. (2019), insufficient information on the aeration system is quite common in research articles. From the review of 21 systems reported in the literature with an average bed depth of 0.53 m, Rous et al. (2019) found no correlation between water depth and aeration efficiency or removal efficiency. Freeman et al. (2018) reported that standard oxygen transfer efficiency was positively related to bed media depth, a result that follows the usually expected trend for biological wastewater treatment systems. However, these results came from column experiments and with bed media depth (between 1.5 and 3.0 m) clearly outside the typical range for CWs. To the best of our knowledge, the influence of bed depth on the efficiency of aerated CWs is a variable for which there is no relevant and mutually (side-by-side) comparable data from real CWs systems, whether on a pilot or field scale.

Therefore, the design and optimization of aeration systems to ensure maximum O_2 transfer from the gaseous to the liquid phase is central to achieving low cost, sustainable treatment solutions with aerated wetlands. Aerated wetland technology is a promising technology for wastewater treatment but data for aerated wetlands are scarce and most of these data are at laboratory scale. This paper attempts to fill these research and knowledge gaps. For this purpose, a demonstration pilot plant was designed and constructed in A Coruña (NW Spain). The configuration consisted of an anaerobic digester used as pre-treatment followed by several HF CWs operated in parallel for raw municipal WW treatment. The effect of bed depth on the treatment efficiency and performance of aerated HF CWs was evaluated.

2. Materials and methods

2.1. Literature search

The Scopus database was searched for scientific literature on aerated CWs with different bed or water depths using the following keywords inserted in "Article title, Abstract, Keywords": "constructed wetlands" OR "treatment wetlands" AND "artificial aeration "OR "aerated wetlands". This search yielded 125 papers, while restricting the search to "depth" reduced the selection to only 12 papers. All 125 paper abstracts were then hand screened, identifying only one paper that analyzed the effect of different medium bed depths, or water depth, on aeration efficiency and CW performance.

2.2. Pilot plant

The pilot plant was built 30 m distance from Faculty of Sciences of University of A Coruña (Spain, UTM coordinates UTM time zone 29, X: 547821.20; Y: 4797233.73). It consisted of a hydrolytic up-flow sludge blanket (HUSB) anaerobic reactor, followed by three horizontal flow constructed wetland units (HFs) in parallel, two of them with forced aeration (HF1 and HF2) and the third one (HFc) without aeration, acting as control. Fig. 1 presents the diagram of the pilot plant design. To cover different working scenarios, effluent recirculation, different aeration modes and different depths were implemented in the HF CWs. Common reed (*Phragmites australis*) was the selected plant species for the wetlands, with a density of 16 plants/m^2, to accelerate the establishment of the plants.

Raw WW was fed by gravity to a grid chamber, fitted with a manual cleaning screening stage. The space between bars of the screening was 5 cm. A bypass for excess flow was also implemented. After the chamber, the influent flowed by gravity to a 1.8 m^3 WW storage tank to supply water for events without influent WW (mainly weekends). The raw WW was collected from the Faculty of Philology, which regularly presented diluted characteristics due to the combined sewage network, which also included rainwater (Pascual et al., 2016). To increase the strength of the influent WW, a concentrated synthetic substrate made off mixture of urea, trisodium phosphate, sodium acetate, starch and municipal primary sludge was added to the storage tank by means of a peristaltic pump model Verdeflex R2S 1CH 3R of 200 L/d, from a 600 L tank.

After the storage tank, a peristaltic pump model Verderflex R6 12.7VP of 3000 L/d was used to feed the HUSB digester. Afterwards, another peristaltic pump model Verdeflex Rapide R3S supplied each aerated bed with the primary treated WW (HF1 and HF2) at a flow rate of 1000 L/d while the control bed was feed by a peristaltic pump model Verdeflex R2S 1CH 3R at a flow rate of 200 L/d. At the end of the process, several effluent collection tanks were placed to store the effluent (one for each HF unit). These tanks were equipped with submergible pumps to recirculate the effluent to the HFs.

The primary treatment, namely a HUSB digester was built with concrete cylinder of 0.70 m in diameter and an active height of 1.8 m, with a total active volume of 0.69 m^3. In this kind of digester, methane production is avoided or minimized (Álvarez et al., 2003), as it only operates in the hydrolytic stage, so separator gas/liquid/solid is not necessary.

All HF CWs have 1.4 m width and 4 m long (surface of 5.6 m^2). Total depth of HF CWs was 0.90 m for HF1 and 0.60 m for the other two (HF2 and HFc), and the water level was set at 0.80 and 0.50 m for the deeper and shallower units respectively. The beds were filled with crushed granitic gravel of 12–16 mm in diameter, with a porosity of 40 %, which means a void volume of 1.79 m^3 for HF1 and 1.12 m^3 for HF2 and HFc. More details of the pilot plant and start-up are described in Pascual et al. (2016).

The Influent manifolds of the beds were made from PVC pipes Ø 200 mm, with 30 mm holes at 25 cm intervals along the pipe, and placed at 30 cm to the bottom of the wetland and embedded in coarse gravel of 50–60 mm in diameter. The effluent pipes manifolds were made in similar way, but placed 15 cm from the bottom of the bed. A PVC pipe with 90° elbow located in an outlet well controlled the water level of each HF unit. The HF effluent discharged to the outlet wells flowed by gravity to the final effluent collection tanks. Fig. 2 shows a construction detail (A, B), plastic chambers during gas emission campaigns (C) and view of the planted units (D).

The aeration system was based on Forced Bed Aeration (FBA®,

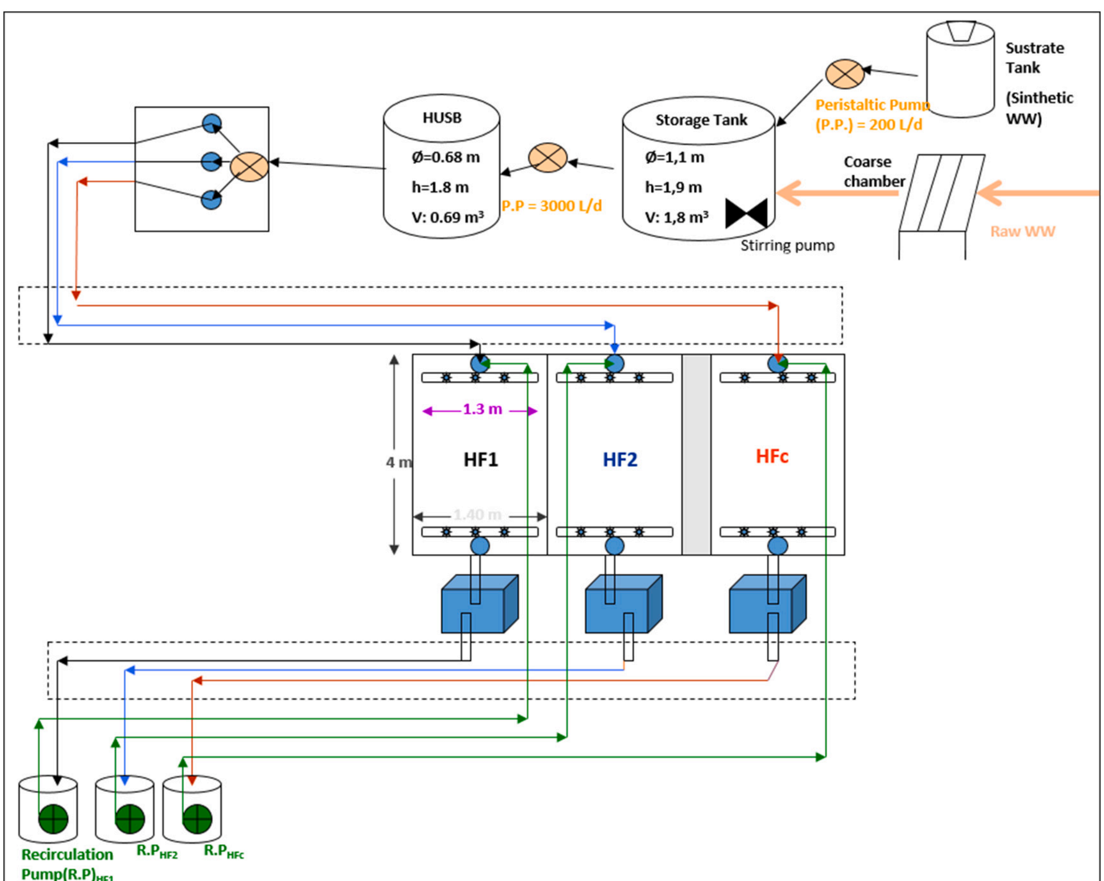


Fig. 1. Schematic diagram of the pilot plant.

Fig. 2. Construction details (A, B), plastic chambers during gas emission campaigns (C) and view of the planted units (D).

developed by Naturally Wallace Consulting, US), and consisted of non-compensated on line drip irrigation pipes placed on the bottom of the beds and pressurized by blowers, to ensure an equal air distribution throughout the wetland. Aeration was provided to all units except HFc. The aeration regime can be adjusted to increase or decrease the oxygen concentration in the WW to degrade the organic matter and the nitrogen compounds.

2.3. Sampling and analysis

Composite influent and effluent samples were collected, over 24 h periods, once or twice a week. An automatic sampler, type 1350 of American Sigma, was used for the influent sampling, while effluent samples were taken in the storage final tanks. The samples were analyzed in the laboratory for Total Suspended Solids (TSS), Dissolved Oxygen Demand (COD), Biological Oxygen Demand (BOD_5), ammonium, total nitrogen (TN), nitrate, nitrite, phosphate and total phosphorus (TP). Temperature, pH, oxidation-reduction potential (ORP) and dissolved oxygen (DO) were determined in situ the same sampling days. An integrated pH & redox 26 Crison electrode was used for pH and ORP determination, a selective electrode (Crison 9663) for ammonium, and an electrode ProODO® from YSI for DO. Electrical conductivity (EC) was measured with a COND600 electrode from Eutech instruments. Anions and cations (NH_4^+, NO_3^-, NO_2^-, PO_4^{3-}, SO_4^{-2}) were determined by ion chromatography (Metrohm 882/863). Inductively coupled plasma optical emission spectrometry (ICP-OES) was used for TP determination. All the analytical methods were carried out as described in Standard Methods (APHA, 2005).

2.4. Operation conditions

To achieve the best nutrient and organic matter removal efficiency, different recirculation and aeration ratios were tested in the pilot plant HF units as indicated in Table 1. The aeration pump used was a diaphragm pump, operating at aeration rate of 64 L/min (0.69 $m^3/m^2 \cdot h$). An aeration regime of 5 h "on" and 3 h "off" were applied during campaigns C1, C6 and C7, while 3 h "on" and 5 h "off" were established during campaigns C2, C3, C4 and C5.

Different recirculation ratios (defined as the ratio between the recirculated flow and the influent flow) from 1 to 1.5 were applied to HF1 and HF2 during campaigns C1 and C2, while HFc had a recirculation ratio of >3. No recirculation was applied during C3, C4 and C5 campaigns. Recirculation ratios of 0.36 and 3 were applied during campaigns C6 and C7, respectively. As shown in Table 1, recirculation was sent directly to the inlet zone of the HF units in all campaigns except during the C6 campaign in which it was directed to the HUSB digester.

Regarding hydraulic loading rate (HLR) ranged from 126 to 199 mm/d to the aerated HF1 and HF2 units, and from 34 to 58 mm/d in HFc. Mean values were 152 + 29 (HF1), 157 + 27 (HF2) and 41 + 8 mm/d (HFc), indicating fairly good uniformity in HLR applied throughout the study. The highest surface loading rates (SLR) occurred during the C3 and C4 campaigns, while the lowest SLRs were achieved

Table 1
Parameters and operation conditions of the pilot plant during the 7 campaigns.

Campaign	C1	C2	C3	C4	C5	C6	C7
Period (days)	8–32	65–95	96–130	131–152	153–170	200–246	247–270
Aeration frequency ON/OFF (h)	5/3	3/5	3/5	3/5	3/5	5/3	5/3
Recirculation ratio (R)							
HF1	1.2	1.5	0	0	0	0.4[a]	2.9
HF2	1.1	1.3	0	0	0	0.4[a]	2.9
HFc	3.7	3.6	0	0	0	0.4[a]	3.0
HLR (mm/d)[b]							
HF1	156.9	126.3	127.9	131.3	138.2	194.3	187.5
HF2	160.1	133.6	133.1	138.0	144.2	199.0	187.5
HFc	38.0	39.5	38.4	40.0	33.8	58.1	37.5
SLR ($gBOD_5/m^2{\cdot}d$)							
HF1	31.9	44.3	76.8	60.7	28.8	20.1	53.5
HF2	32.6	46.9	79.9	63.5	30.1	20.6	53.5
HFc	7.7	13.9	23.0	16.4	7.8	6.0	10.2
SLR ($gTN/m^2{\cdot}d$)							
HF1	6.1	5.3	5.38	6.42	4.35	3.7	4.6
HF2	6.3	5.6	5.59	6.72	4.55	3.8	4.6
HFc	1.5	1.7	1.61	1.73	1.18	1.1	0.9

[a] Recirculation of HF effluent to the HUSB inlet.
[b] Recirculation was not included in HLR calculation (To obtain the overall hydraulic loading rate, including recirculation, the given values must be multiplied by the factor (1 + R)).

during C6. In the aerated units, the SLR ranged from 20 to 80 g $BOD_5/m^2{\cdot}d$ and from 3.7 to 6.7 g $TN/m^2{\cdot}d$, while they ranged from 6 to 23 g $BOD_5/m^2{\cdot}d$ and from 1.1 to 1.7 g $TN/m^2{\cdot}d$ in the control unit. These SLR in the control unit were selected in order to match the reference values for subsurface HF CWs in the literature (Suhad et al., 2018) while it was hypothesized that aerated units could operate efficiently with SLRs up to 3 to 5 times higher. Both the HF1 and HF2 aerated units received very similar HLRs and SLRs, which ultimately were only 3 % higher on average for the HF2 than the HF1 unit.

In addition to SLR, volumetric loading rate (VLR) will be used to analyze the results. The VLR represents the organic loading rate per unit volume (liquid or void volume) of the system. Both parameters are related by the following equation:

$$VLR = SLR/(H{\cdot}e)$$

where H is the bed height (to the water level) and e is the bed porosity.

The hydraulic retention time (HRT) can be calculated for each CW unit and operating period by applying the following equation:

$$HRT = (H{\cdot}e)/HLR$$

The calculated mean HRT values were 2.17 ± 0.37 d (HF1), 1.31 ± 0.21 d (HF2) and 5.03 ± 0.76 d (HFc). From the duration of the operation periods listed in Table 1, we obtain that they included on average 6.0 times HRT (HFc), 22.3 (HF2), or 13.5 (HF1). With the sole exception of the C5 campaign of HFc, these values were higher than the usual criterion of 3–4 times TRH to achieve a pseudo-steady state in the functioning of biological systems.

2.5. Biomass sampling, biological assays and gas emission measurements

Four mesh steel cylinders Ø 20 cm were installed in all HFs, two at 80 cm from the inlet (left and right side, at 35 cm from the wall) and two at 80 cm from the outlet (left and right side). Four more cylinders of plastic mesh with 8 cm diameter were inserted in the steel cylinders to evaluate biomass production (Fig. 2A, B). The cylinders reach the bottom go the bed since they had the same height as the bed depth. All the cylinders were filled with the same gravel used as media for the beds. At the end of the sampling campaigns, the internal cylinders were manually extracted, to determine the amount of biomass accumulated and its characteristics.

Aliquots of 80 g of gravel from the top and bottom half parts of each cylinder were used to determine total solids (TS) content by drying at 105 °C up to constant weight and then calcined to 550 °C for volatile solids (VS) content. Samples of approximately 100 g of wet gravel integrated from the content of each cylinder were used for biological assays to measure the methanogenic, nitrifying and denitrifying activity of the accumulated biomass. Biological assays were incubated in 250 mL bottles. Nitrification assays were fed with 150 mL of a solution (50 mg NH_4^+-N/L and 1000 mg $NaHCO_3$/L), while denitrification assays were fed with 100 mL of a solution (50 mg NO_3-N/L and 500 mg COD/L of sodium acetate). Methanogenic assays were fed with 100 mL of a solution containing 500 mg COD/L of sodium acetate. Macro and micronutrients were added in all assays at a ratio of 1 mL/L of the stock solutions as suggested by Carballeira et al. (2017). $Na_2S{\cdot}xH_2O$ (100 mg S/L) was added to anaerobic and anoxic assays. All assays were carried out in duplicate and incubated at 20 °C in a thermostatic chamber.

Nitrate concentration was monitored to determine its production or removal rate. The nitrification and denitrification volumetric rate was obtained as the slope of the curve plotting of nitrate concentration versus assay time, once the lag phase was overcome. This volumetric rate was then converted to specific rate per unit of TS, given the values of the specific nitrifying activity (SNA) and specific denitrifying activity (SDA), respectively from the nitrification and denitrification assays. Methanogenic assays were monitored following the head-space gas analysis method (Carballeira et al., 2017). Methane production rate was obtained from the slope of the cumulative methane production curve versus time and then converted to specific methanogenic production rate per unit of TS, given the values of the specific methanogenic activity (SMA). The potential methane emission (PME, mg $CH_4/m^2{\cdot}d$) was obtained as the product of SMA by the TS content per unit area of each CW.

In order to measure greenhouse gas (GHG) emission rates, the emitted gases from the wetland surface were collected in airtight plastic chambers of 25 L of volume placed inverted against the wetland surface (Fig. 2C). To ensure a water seal isolated the chambers from the external atmosphere, the chambers were buried on the gravel media until reaching the water level. Two chambers were installed in each of the HF unit during the campaigns C3 and C6, one close to the inlet and the other close to the outlet. Gas from the chambers was sampled with a syringe at different time intervals. Methane, nitrous oxide and carbon dioxide content were determined by gas chromatography with thermal conductivity detector (GC-TCD). The surface emission rate (SER) and emission factors (EF) of each gas were obtained from the evolution of the percentage of that gas in the confined headspace, following the methods described by de la Varga et al. (2015). To estimate the methane emission factor (EF, as the percentage of CH_4-C emitted to fed TOC), a BOD_5 to TOC conversion factor of 0.5 g TOC/g BOD_5 was used (Mander et al., 2014).

2.6. Statistical analysis

Statistical analysis and calculations were performed using an Excel program. The forward stepwise method was used to build the multilinear regression models (Navidi, 2006). The coefficient of determination (R^2), statistical F-value, and probability (p) were used to assess the adequacy of the least-squares fit. Data sets from parallel experiments were compared using one-way and two-way analysis of variance.

3. Results

3.1. Characteristics of the influent WW to the HFs units

Seven sampling campaigns (C1 to C7) were developed during the research period. Table 2 presents the influent concentrations of WW to the HF units, after the concentrate addition and pre-treatment in the HUSB digester. Total solids removal was close to 72 % in the HUSB digester for the period, which can be considered as an efficient pre-treatment to avoid solids reaching the wetlands and consequently preventing clogging of the systems (de la Varga et al., 2013). During C3 and C4 campaigns, the influent concentration of TSS was higher due to the accumulation of suspended solids in the HUSB digester that were released to the systems. It was solved by increasing the excess sludge purge frequency from the HUSB digester. Regarding the other water quality parameters analyzed, the average influent concentration throughout the study for BOD_5 ranged from 104 (campaign C6) to 600 (C4) mg BOD_5/L, For TN the concentration ranged from 19.5 (C6) to 45.5 (C4) mg TN/L and for TP, the range was from 12.2 (C4) to 23.5 (C4) mg TP/L (Table 2). Thus, in general, the C3 and C4 campaigns showed the most concentrated influent and the C6 campaign the lowest. COD concentration followed BOD_5 concentration (Table 2), showing a strong correlation among these two parameters (R^2 = 0.946; p = 0.000). The influent to HF units showed an average BOD_5/COD ratio of 0.63 ± 0.09.

Table 2
Concentration of influent to HF units.

Campaign	C1	C2	C3	C4	C5	C6	C7
pH	7.3 ± 0.9	7.5 ± 0.3	6.4 ± 0.6	7.1 ± 1.2	8.2 ± 0.9	8.2 ± 0.7	7.2 ± 0.6
EC (μS/cm)	755 ± 303	989 ± 158	995 ± 132	1112 ± 33	854 ± 134	625 ± 117	802 ± 75
SS (mg/L)	137 ± 89	71 ± 18	192 ± 142	141 ± 86	58 ± 8	55 ± 27	64 ± 21
BOD_5 (mg/L)	203 ± 113	351 ± 81	600 ± 83	449 ± 218	213 ± 43	104 ± 48	285 ± 57
COD (mg/L)	356 ± 202	468 ± 111	847 ± 93	779 ± 261	339 ± 63	207 ± 99	429 ± 62
TN (mg/L)	39.0 ± 19.7	41.6 ± 11.8	42.3 ± 11.4	47.5 ± 7.6	32.2 ± 8.9	19.5 ± 4.6	24.6 ± 3.4
COD/TN	9.1	11.3	20.0	16.4	10.5	10.6	17.4
NH_4^+-N (mg/L)	27.9 ± 14.4	35.2 ± 10.2	29.5 ± 11.7	35.1 ± 0.8	25.4 ± 8.8	14.1 ± 8.4	27,7 ± 9.6
TP (mg/L)	*n.a.*	18.3 ± 8.8	19.1 ± 4.4	23.5 ± 1.3	18.2 ± 3.1	12.2 ± 3.1	13.0 ± 3.2
SO_4 (mg/L)	13.7 ± 1.5	8.7 ± 0.5	7.9 ± 1.6	11.2 ± 0.9	11.7 ± 0.4	9.9 ± 2.2	4.2 ± 2.4
T (°C)	14.2 ± 1.3	19.0 ± 1.8	20.9 ± 1.7	24.2 ± 0.5	25.4 ± 0.6	22.8 ± 1.3	*n.a.*

Mean influent concentration of NO_3-N and NO_2-N were 0.13 and 0.07 mg/L, respectively, whilst punctual maximum values reached 3.4 (campaign C6) and 0.7 (C1) mg/L, respectively. Mean influent concentration of dissolved oxygen was 1.2 ± 1.1 and mean oxidation-reduction potential was −91 ± 90 mV. n.a.: not available. For parameter acronyms, see Section 2.3.

3.2. Effluent concentration of HF units

Fig. 3 shows the evolution of the concentration for the different parameters in the effluent of the HF units. Effluent pH ranged from 6.8 to 8.2 for the overall operation period, while mean values during the campaigns ranged from 7.4 to7.7. Considering mean values from campaigns, effluent pH was 7.6 ± 0.1 for units HF1 and HF2 and 7.4 ± 0.2 for unit HFc. ORP showed very similar values in the three units, being partially correlated with DO concentration and particularly with the application of recirculation. Higher ORP and DO values were observed during periods C1 and C6. It seems that the recirculation of effluent increased ORP in all units (Fig. 3). While the mean temperature for the influent was 21.1 ± 4.1 °C, the effluent of HF units showed temperatures of about 4 °C lower: 17.7 ± 4.1, 17.6 ± 4.1 and 16.8 ± 3.6 °C for HF1, HF2 and HFc effluents, respectively (data not shown).

Effluent COD concentrations were generally under 50 mg COD/L for HF1 and HF2, except for HF2 during C3 and C4 (the campaigns with the highest organic load rates). HFc showed also COD concentrations below 50 mg/L, except during the first 50 days of operation, possibly due to a slower start up process in this unit than in the aerated ones. BOD_5 and TSS in the effluents were close to zero in most of the samples (data not shown). Thus, regarding organic matter removal, the control unit and the aerated HF1 unit showed sufficient capacity to deal with the applied organic loading rate and reach a high quality effluent. In contrast, the aerated unit HF2, with a lower bed depth than the HF1 unit, showed a more limited capacity as indicated by COD (and BOD_5) accumulation in the effluent during the higher loading periods C3 and C4 (Fig. 3).

Ammonia and nitrate concentration in the effluent of the different units performed distinctly, depending on factors such as aeration intensity, recirculation and loading rate. The effluent from the control unit showed g the highest ammonia concentration, being only surpassed by the HF2 effluent in the overloaded periods C3 and C4, as well as during campaign C5 (Fig. 3). The nitrification capacity in HFc was limited, except for campaign C5 and the first days of campaign C6 (Fig. 3) when the ammonia concentration in the effluent sharply decreased. This decrease was accompanied by an increase of nitrate concentration, which accumulate in the effluent. Ammonia removal and nitrate accumulation in HFc unit during this period could be due to the large decrease of the influent BOD_5 concentration and applied SLR as well as the decrease in the COD/TN ratio (Table 2). In these conditions, the oxygen transfer rate in the non-aerated unit will be sufficient to promote nitrification and limit denitrification. This period corresponded to the months of June and July, when the macrophytes were fully developed and in the highest vegetative state. The effect of these factors in decreasing the effluent ammonia concentration was also observed for HF1 and HF2 units (Fig. 3).

The aerated units reached very low effluent concentrations of ammonia during periods C1, C6 and C7 in which a larger period of aeration was maintained (aeration regime of 5 h on and 3 h of, Table 2). Mean effluent concentration during these periods were 0.1 ± 0.2 and 1.2 ± 2.5 mg NH_4^+-N/L for HF1 and HF2 units, respectively. However, during the periods with reduced aeration to 3 h on and 5 h off (C2, C3,

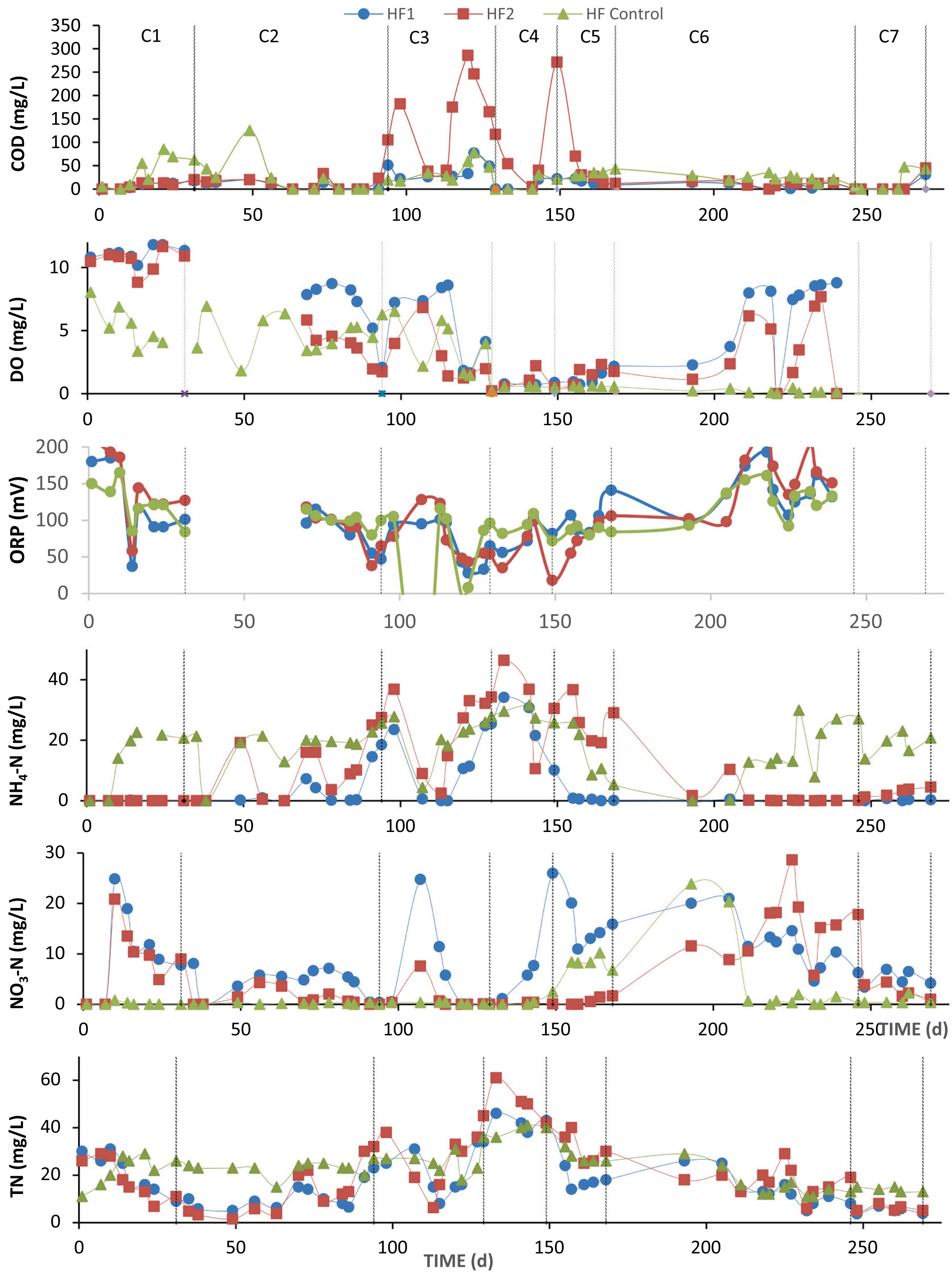


Fig. 3. Evolution of the concentration for different parameters in the effluent of the HF units and evolution of ORP. Dashed lines indicate the separation between campaigns.

C4 and C5), effluent ammonia concentrations increased to 10.0 ± 11.3 (HF1) and 23.0 ± 11.9 mg NH_4^+-N/L (HF2) on average. As reference, the HFc unit showed mean effluent ammonia concentrations of 18.0 ± 7.1 (C1, C6 and C7) and 20.9 ± 7.3 mg NH_4-N/L (C2-C5).

These results show that, depending on the loading rate applied, the aerated units can reach close to complete ammonia removal if the aeration period was conveniently extended and calculated according to the influent concentrations. In fact, the deeper aerated unit HF1 reached very low ammonia concentration (0.38 ± 0.34 mg NH_4^+-N/L, $n = 5$, Fig. 3) during period C5 with an aeration regime of 3 h/5 h on/off and low SLR of approximately 29 g $BOD_5/m^2 \cdot d$ and 4.4 g $TN/m^2 \cdot d$ (Table 1). Very low ammonia effluent concentration was also reached by HF1 unit during period C7 with an aeration regime of 5 h/3 h on/off and higher SLR of approximately 54 g $BOD_5/m^2 \cdot d$ and 4.6 g $TN/m^2 \cdot d$. However, at SLR of 61–77 g $BOD_5/m^2 \cdot d$ and 5.4–6.4 g $TN/m^2 \cdot d$ (periods C3 and C4), the aeration regime of 3 h/5 h on/off was not sufficient and the effluent concentration was above the discharge limit of 10 mg NH_4^+-N/L (Fig. 3). The unit HF2 with only 0.5 m water table depth clearly showed worse ammonia removal capacity than the HF1 unit with 0.8 m water table depth.

Effluent concentration of nitrate was higher in HF1 unit (9.9 ± 4.4 mg NO_3-N/L) than in HF2 (5.0 ± 6.8 mg NO_3-N/L) and HFc (2.0 ± 3.2 mg NO_3-N/L) units. Nitrate concentration was higher in the effluent of HF1 respect to HF2 unit in all campaigns except in C6 in which the opposite occurred (Fig. 3). Finally, the evolution of TN concentration in the effluents showed little differences among units, although the variation over time appeared to be smaller in HF1 compared to HF2.

3.3. Efficiency of pollutant removal and loading rate

3.3.1. Organic matter removal

Fig. 4 shows the percentage removal of COD plotted against influent concentration, surface loading rate and volumetric loading rate. The statistical data for correlations presented in Fig. 4 are given in Table 3. BOD_5 was selected as the reference parameter for this analysis because it is an accurate indicator of the loading rate of biodegradable matter. However, as previously indicated, a correlation between influent COD and BOD_5 concentrations existed, thus indicating that similar results would be obtained for the case of using COD instead of BOD_5. On the other hand, COD removal was selected as the efficiency parameter, because it showed a clear variability among periods, while TSS and BOD_5 were found to be near 100 % removal in most cases. In addition, the inclusion of several independent variables (BOD_5 concentration, SLR, VLR…) for the multivariable regression did not improve the results, since the second variables never reached statistical significance (*p* value).

All the correlations in Fig. 4 have a clear physical meaning, although they are not always statistically significant. In the case of HF1, this is due to the small variation in the dependent variable, which reflects the reduced effect of influent concentration, SLR and HLR on COD removal in this unit. The greater effect of the independent variables on HF2 treatment efficiency makes the correlations significant for this unit. The same happens in some cases for HFc.

Percentage COD removal was always higher in the deeper HF1 than in the HF2 or the HFc. Regarding the effect of the influent BOD_5 concentration (Fig. 4A), the percentage COD removal in HFc unit clearly increased as the BOD_5 increased from 100 to 300 mg /L. This effect was smaller in the HF1 unit where the percentage COD removal was always up to 92 % regardless of the inlet BOD concentration. In the HF2 unit, the contrary effect was observed, probably because SLR and BOD_5 concentration are correlated (because the HLR applied was relatively constant, Table 2), and the efficiency of this unit was more affected by the applied SLR.

Table 3
Statistical data for correlations in Fig. 4.

Unit	N	Variable x	Equation	R^2	p
HF1	7	BOD_5 influent (mg/L)	$y = -3E\text{-}05 \times {}^2 + 0.0225\times + 93.841$	0.434	>0.22
HF2	6	BOD_5 influent (mg/L)	$y = -0{,}0277\times + 99{,}487$	0.867	0.007
HFc	7	BOD_5 influent (mg/L)	$y = -7E\text{-}05 \times {}^2 + 0{,}0725\times + 79.674$	0.626	>0.16
HF1	7	SLR (g BOD_5/m^2·d)	$y = -0.0026 \times {}^2 + 0.2625\times + 91.417$	0.585	>0.12
HF2	6	SLR (g BOD_5/m^2·d)	$y = -0.2216\times + 100.72$	0.836	0.011
HFc	7	SLR (g BOD_5/m^2·d)	$y = -0.093 \times {}^2 + 3.2248\times + 70.946$	0.791	<0.07
All	16	SLR (g BOD_5/m^2·d)	$y = -0.0032 \times {}^2 + 0.1768\times + 94.675$	0.368	>0.20
All	16	VLR (g BOD_5/m^3·d)	$y = -0.0003 \times {}^2 + 0.0517\times + 94.958$	0.886	<0.04

The control unit achieved nearly similar COD removal in terms of percentage, since it operated with an SLR (g BOD_5/m^2·d) four times lower than aerated CWs (Fig. 4B) suggesting that can cope with higher loading since a breakpoint was not reached. For HF1 the best result (99 % COD removal) was achieved for a SLR of 61 g BOD_5/m^2·d. Even with SLR close to 80 g BOD_5/m^2·d, a 96 % COD removal is reached. For HF2, a 91–97 % COD removal took place at a SLR of 20–40 g BOD_5/m^2d, but dropping to 82 % COD removal at SLR of 80 g BOD_5/m^2·d. COD removal for HFc ranged from 86 to 99 % at SLR of 8–21 g BOD_5/m^2·d, which are expected values for conventional non-aerated HF CWs treating domestic WW (Vymazal, 2010). COD removal started to drop for HF1 and HFc at SLR of 80 and 20 g BOD/m^2·d, suggesting possible optimum ranges of application and a factor four times higher for the aerated wetland. On the other hand, COD removal for HF2 decreased when SLR is higher than 40 g BOD_5/m^2·d, which means a factor two times higher than HFc and two times lower than the deeper aerated wetland. This concurs with other studies, such as Aguilar et al., 2021 who report that aerated systems achieved high COD and BOD_5 elimination rates (90 %) at the end of the 5-month test period.

The correlation of COD removal with volumetric loading rate gives a different view (Fig. 4C). The relative ranges of VLR are different from those of SLR. Thus, while the HF2 unit achieved VLR 4 times greater than the HFc unit, the HF1 unit showed VLR only 2 times greater than HFc and 2 times less than HF2. Nevertheless, it is worth noting that the VLR appears as a factor that controls efficiency better than the SLR,

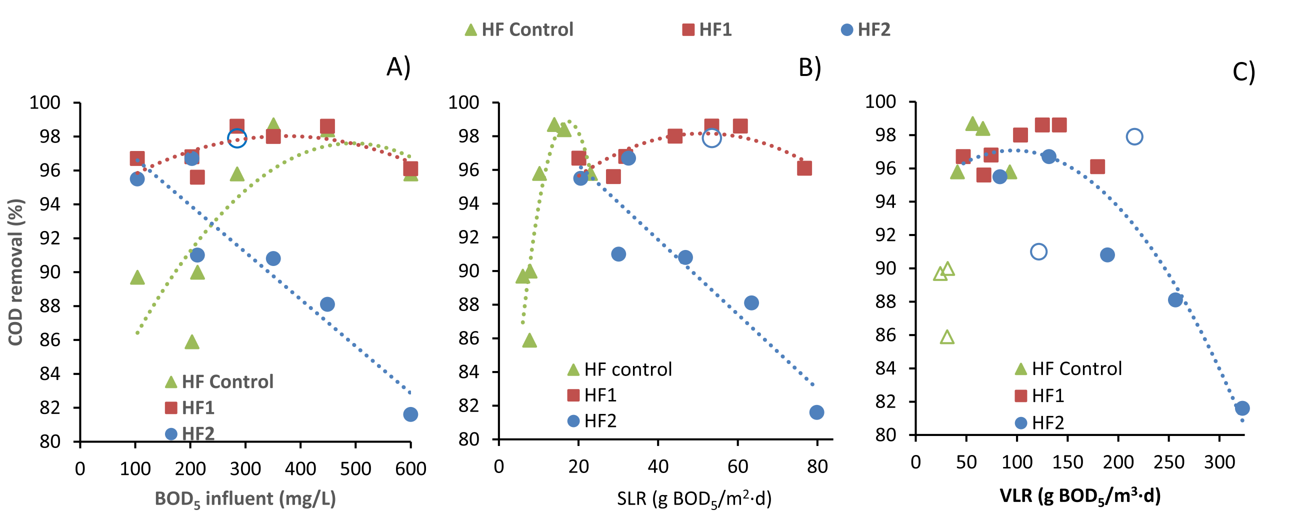


Fig. 4. Percentage COD removal vs influent BOD_5 concentration (A); percentage COD removal vs BOD_5 SLR (B); percentage COD removal vs BOD_5 VLR (C). Non stuffed points are excluded from the equations; these corresponds to C7 from HF2 (A and B), while in C) were excluded the points C5 and C7 from HF2 and C1, C5 and C6 from HFc.

because most of the data from the three units can be modelled using a single correlation equation (Fig. 4C, Table 3).

Comparing HFc vs aerated wetlands, the intensified systems a) perform better at low influent concentrations, which may be due to a higher biodegradation of pollutants in aerobic conditions and higher mass transfer rates favored by the mixing effect of aeration, b) at medium-high concentrations, they can afford approximately double VLR (up to 200 g BOD_5/m^3•d) than conventional HF (up to 100 g BOD_5/m^3•d). In addition, as already mentioned, with aeration, the SLR can be increased by a factor of 4 if the bed depth increases from 0.5 m to 0.8 m, but only by a factor of 2 if the depth remains the same.

3.3.2. Ammonia and total nitrogen removal

Fig. 5 shows the efficiency in ammonia and TN removal for all campaigns in the aerated and control HF units. Elimination percentages were calculated for the average input and output values. Ammonia removal was very high (>99 %) in HF1 during aeration mode 5 h/3 h on/off (campaigns C1, C6 and C7). Although somewhat lower than HF1, HF2 also reached ammonia removals higher than 89 % during these periods. During aeration mode 3 h/5 h on/off, both units showed lower nitrification efficiency, which decreased as the loading rate increase during campaigns C2 to C4 (Fig. 5A). Again, the percentages of ammonia removal were lower for the HF2 than for HF1 unit. The greater difference between ammonia removal in units HF1 and HF2 was observed during period C5, which was a transient period of low loading rate after the preceding periods of high loading rate (Table 1). This performance registered suggests a higher adaptation capacity of the deeper unit HF1 in comparison to the shallower HF2. The control unit showed always lower ammonia removal efficiency than the aerated units (Fig. 5A). In overall, ammonia removal reached 80 % (HF1), 52 % (HF2) and 27 % (HFc) on average.

TN removal was generally higher in HF1 (44,7 % on average for the entire operation period) while, depending on the conditions, HF2 and HFc showed similar TN removals, reaching 33 % and 34 % TN removal on average, respectively (Fig. 5B). Best results were achieved for all the wetlands during campaign C7, when the highest recirculation ratio of 2.9 (HF1, HF2) and 3.0 (HFc) was applied. According to Gonzalo et al. (2017), effluent recirculation favored TN as shown in Fig. 5B for periods C1, C2 and C7 which operated with recirculation rates ranging from 1.2 to 3.0 (Table 1) and reached TN removal 53 % to 79.% in the aerated units (HF1 and HF2) and from 36 % to 43 % in the control HFc unit. On the contrary, during periods with no recirculation to the HF units (C3 to C6), TN removal was clearly lower, ranging from 11.1 % to 47.4 % for HF1 and from nearly zero to 38 % for HF2 and HFc. The highly variable

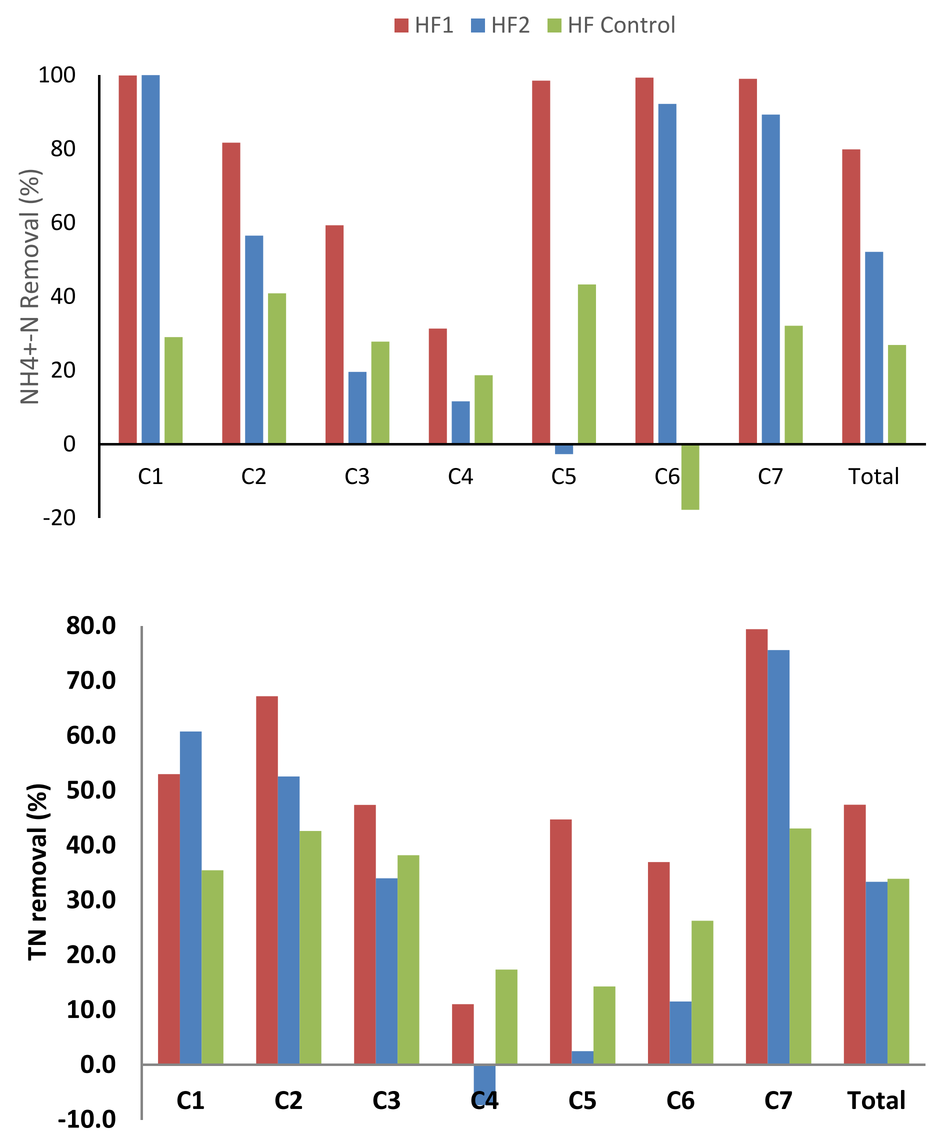


Fig. 5. Ammonia and TN removal for all campaigns in the HFs (Percent removal values obtained from mean influent and effluent values for each period given in Tables S1, S2, and S3, see Supplementary material).

TN removal during non-recirculation conditions was due to the variation of the loading rate and the COD/TN ratio (Tables 1 and 2). These results obtained are in agreement with those obtained by Aguilar et al., 2022 who reported higher nitrate and TN removal (52.8 % and 46.8 %, respectively) during operation under aerated-conditions.

Considering mass removal rates, during periods with 5 h/3 h on/off aeration, C1 and C7, both aerated units reached similar surface nitrification rates (4.8 ± 0.5 and 4.5 ± 0.1 g NH_4^+-N/m^2·d for HF1 and HF2, respectively). On the other hand, the volumetric nitrification rate was clearly higher in the HF2 unit (18.4 ± 0.5 g NH_4^+-N/m^2·d) compared to the HF1 (11.1 ± 1.3 g NH_4^+-N/m^2·d). TN removal followed similar behavior, being 0.73 (HF1) and 0.80 (HF2) times the nitrification rate.

Fig. 6 shows the influence of BOD_5 and TN loading rate on ammonia removal in the three HF units. In order to present the main trends, some scattered values of ammonia removal were excluded from the correlation equations. These values corresponded to negative ammonia removals showed by HF2 during period C4 and by HFc during period C6. High values of ammonia removal (near 100 %) in HF1 and HF2 units during period C1 were also excluded from correlations with TN loading rate.

As can be seen by comparing Figs. 6 and 4, the decrease of the removal percentage with the increase in the loading rate was higher for ammonia removal than for COD removal. While COD removal efficiency in the aerated units was better described by VLR of BOD_5 than by SLR of BOD_5 (Fig. 4A and B), for nitrification efficiency the difference among the effect of these two loading rate parameters was not clear (Fig. 6A and B). This is indicated by the correlation lines showing similar slopes and efficiency reductions in similar ranges of SLR of BOD_5. In addition, when considering the TN loading rate as the control parameter of ammonia removal, Fig. 6C and D suggest that was the SLR and not the VLR the parameter that better described the nitrification efficiency. In this way, while two separate correlation equations for HF1 and HF2 are required to accurately describe NH_4^+-N removal by TN VLR, a combined unique equation using TN SLR can represent the nitrification efficiency. Fig. 6D indicates that similar NH_4^+-N removal efficiency can be obtained in HF2 and HF1 units at VLR approximately 50 % higher in the shallower HF2 unit than in the deeper one. However, a different effect of TN SLR on NH_4^+-N removal also exist (Fig. 6C), showing the HF1 and HF2 units had similar efficiency when a 25 % lower SLR was applied in the shallower HF2 unit than in HF1. Finally, Fig. 6 also shows that HFc operated at distinctly lower SLR or VLR and lower NH_4^+-N removal efficiency than the aerated units did, but without a significant trend with the loading rate. For ammonia removals similar to those obtained in the HFc unit, which are generally low, the aerated units supported SLRs at least 5 times higher than the non-aerated unit did.

3.4. Biomass accumulation and microbial activities

3.4.1. Biomass concentration and distribution

Biomass accumulation in wetlands and moisture retention is shown in Fig. 7. Biomass content, in terms of VS per unit of TS of gravel bed, was very similar in the three units, which showed mean values of 0.42 ± 0.10 (HF1), 0.44 ± 0.17 (HF2) and 0.40 ± 0.05 %VS (HFc), without significant differences among units ($p > 0.45$).

Regarding HF1, biomass accumulation was significantly higher in the inlet part than in the outlet part of the wetland ($p = 000$). The biomass is concentrated at the beginning of the unit probably due to the higher organic loading at the inlet of the system as well as the sedimentation and filtration solids processes that occur at the beginning of the bed. On the other hand, there is no significant difference of biomass accumulation between bottom and top part of the gravel bed and the right and left side of the wetland ($p > 0.8$), which suggest a very good hydraulic distribution of the WW through the gravel bed.

In the case of HF2, the tendency is similar to in HF1, thus showing a higher biomass concentration in the inlet zone than in the outlet zone of the unit ($p = 0.025$). Although biomass distribution between the bottom and the top zones and right and left sides was variable (Fig. 7), differences were not significant ($p > 0.3$). The biomass accumulation in HFc was more uniform among the various sampling zones and significant differences were not found ($p > 0.13$) for bottom/top or right/left distribution.

A higher content in biomass led to a higher moisture retention after

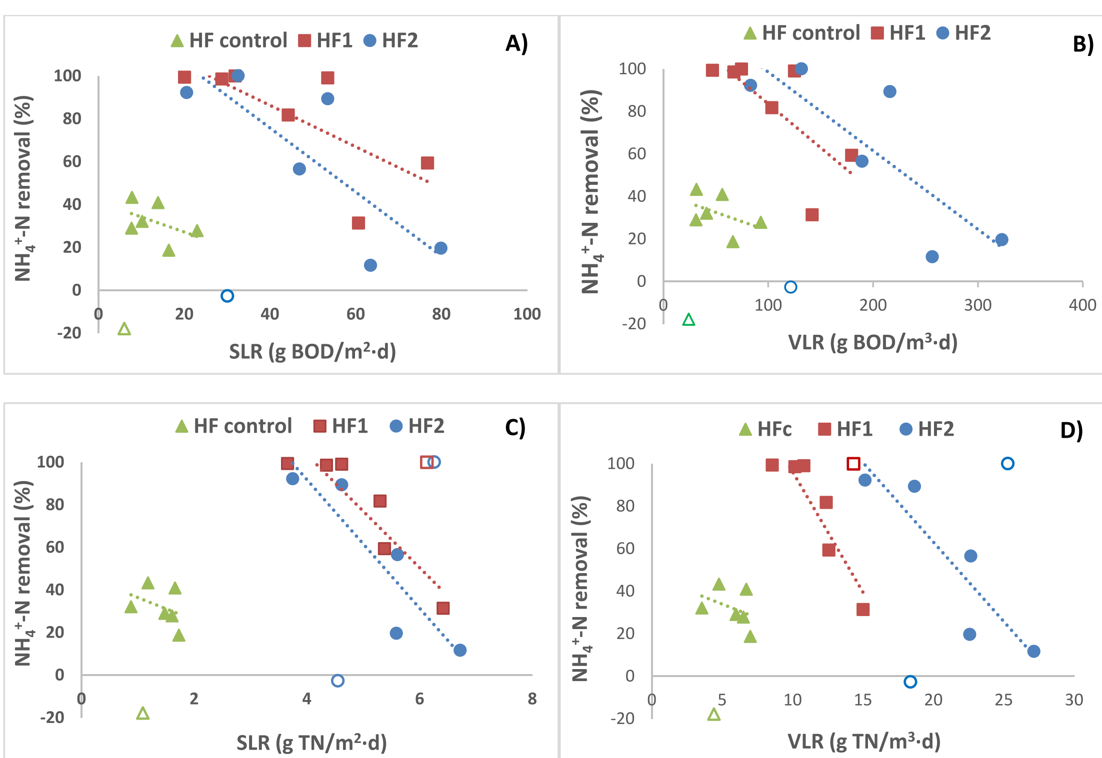


Fig. 6. Correlation of NH_4^+-N removal with several loading rate parameters (non stuffed points are excluded from the equations).

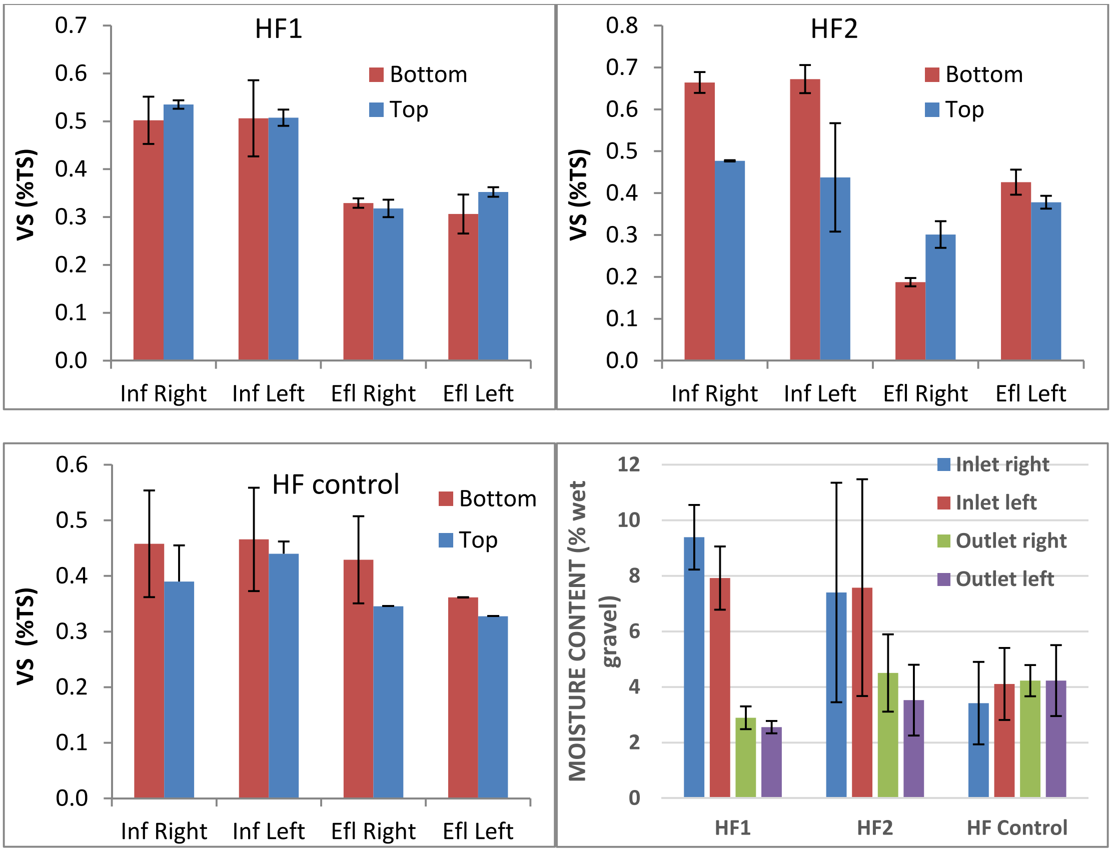


Fig. 7. Biomass accumulation in the different parts of the wetlands, expressed as volatile solids adhered to the gravel, and moisture retention.

draining the bed, as indicated by the linear correlations found between both parameters for HF1 ($R^2 = 0.91$) and HF2 units ($R^2 = 0.55$). The correlation was poor for the HFc unit because of the reduced variation in both parameters.

3.4.2. *Specific nitrifying and denitrifying activities*

Table 4 presents the nitrifying denitrifying obtained for the different zones of each HF unit. For the control HFc unit, the SNA was higher in the outlet than in the inlet part of the bed. This could be due to a lower organic matter content in the outlet part than in the inlet zone, which made residual oxygen available for nitrifying microorganisms. In addition, nitrification assays with HFc biomass showed a high lag phase of about 46-48 h in all wetland position (data not shown). SNA in HFc was slightly higher than the values of 0.30 mg N/kg TS•h obtained by Torrijos et al. (2018) in lab scale mesocosm simulating horizontal constructed wetlands. Lv et al. (2013) reported SNA values of 0.25 and 0.19 mg N/kg TS•h for planted and unplanted, respectively, non-aerated lab-scale HF CW. In the present study, higher SNA values were obtained for the aerated units than for the non-aerated control unit. For HF1, SNA was slightly higher in the inlet part than in the outlet part, but the lag phase in assays was similar in both parts of the wetland (20−22*h*). For HF2, nitrifying activity was much higher in the inlet part than in the outlet part. The lag phase was only 2–3 h at any position of the HF2 unit. The higher SNR in HF2 compared to HF1 agrees with the respective TN SLR and ammonia removal rates in Fig. 6.

Table 4
Specific nitrifying activity (SNA) and denitrifying activity (SDA) in the different parts of the wetlands.

Position	SNA (mg NO_3-N/kg TS·h)			SDA (mg NO_3-N/kg TS·h)		
	HF1	HF2	HFc	HF1	HF2	HFc
Inlet right	1.14 ± 0.18	3.27 ± 0.14	0.39 ± 0.11	4.96 ± 0.37	16.14 ± 1.66	2.20 ± 0.10
Inlet left	1.01 ± 0.01	2.39 ± 0.22	0.41 ± 0.04	6.06 ± 0.62	16.20 ± 4.41	2.47 ± 0.23
Outlet right	0.76 ± 0.04	0.71 ± 0.06	0.60 ± 0.19	4.78 ± 0.25	2.37 ± 0.35	2.09 ± 0.09
Outlet left	0.65 ± 0.08	0.20 ± 0.11	0.53 ± 0.03	2.46 ± 0.45	3.05 ± 0.47	2.20 ± 0.21
Mean	0.89 ± 0.23	1.64 ± 1.43	0.48 ± 0.10	4.56 ± 1.51	9.44 ± 7.78	2.24 ± 0.16

Regarding SDA, the HFc unit showed similar values in the inlet and outlet parts of the unit, while the lag phase was low in both positions (8–12 h). The results were in the range of those previously reported by Lv et al. (2013) and Torrijos et al. (2018), which ranged from 1.2 to 5.5 mg N/kg TS•h. Higher SDA values were obtained in the present study for the aerated units (Table 4). The assays showed that the denitrification process was more developed in the initial part of the HF1 unit where the lag phase was significantly lower (nearly zero) than in the outlet part (20*h*). For HF2, the SDA at the inlet part was almost 3 times higher than in HF1, while at the outlet part was similar. However, the lag phase was low in both positions of the HF2 unit. The high SDA in aerated units indicate that denitrifying bacteria can survive in alternating aerobic/anaerobic environments (Redmond, 2012), which resulted in faster denitrification rates in aerated wetlands, during the periods when the anaerobic conditions prevail. Murphy et al. (2016) also reported in a mature aerated wetland that nitrification recovery occurs in a matter of hours after re-starting aeration.

The comparison between specific activities in the biomass from aerated and non-aerated units indicates that intermittent aeration greatly increased both nitrifying and denitrifying activities in the wetland inlet zone, while differences were reduced in the outlet zone (Table 4). The increase observed in the inlet zone was higher for the HF2 unit than for the HF1 unit. In this way, compared to HFc, mean values of both nitrifying and denitrifying activities were approximately 2 times higher (HF1) and 4 times higher (HF2).

On the other hand, the nitrifying/denitrifying activities ratio was constant along all units and unit position, being 0.20 ± 0.07 on average. Thus, the denitrifying activity was approximately 5 times higher than the nitrifying activity in all HF units. This suggests that nitrifying/denitrifying activities ratio is not affected by aeration or by other design criteria such as the height or the HLR applied (Torrijos et al., 2018). Therefore, when oxygen concentration is limited, the denitrification process is complete, while in good aeration conditions the existing denitrification potential is limited by unfavorable conditions. Therefore, correctly controlling the aeration period, that is, the relative duration and distribution of on/off aeration periods, the total nitrogen removal can be optimized to improve nitrification and denitrification process together with TN removal.

The specific biomass activities were obtained in non-limiting substrate conditions, then representing maximum activities. Potential maximum nitrification and denitrification rates can be obtained multiplying the specific activities by the total amount of TS per square meter (or void cubic meter) in the gravel bed. The potential rates can be compared with actual removal rates. The calculations indicate that only 1.2 % of the potential denitrifying rate and 4.7 % of the potential nitrifying rate in the control unit (HFc) were used during the campaign C7 (the one that corresponded to the moment when biomass concentration and activities were determined). However, the percentage used of the potential denitrifying rate increased to 2.6 % (HF2) and 3.6 % (HF1) in the aerated units. The percentage used of the potential nitrifying rate also increased to 19.9 % (HF2) and 23.5 % (HF1). These figures mean that the efficiency in the use of the potential microbial activities increase by a factor of 2 (denitrifying) and 4 (nitrifying) in the shallow aerated bed with respect to the unaerated bed, and by a factor 3 (denitrifying) and 5 (nitrifying) in the case of the deeper aerated bed. This increased efficiency can be attributed to enhanced mass transfer rate favored by the mixing effect of aeration (nitrifying and denitrifying), as well as by the greater availability of oxygen as substrate (nitrifying).

3.4.3. *Specific methanogenic activity and greenhouse gas emissions*

Obtained values of specific methanogenic activity in anaerobic and anoxic conditions are shown in Table 5. SMA in anaerobic conditions was similar in the three units ($p = 0.34$) and clearly decreased from the inlet zone to the outlet zone ($p = 0.005$). In anoxic conditions, the SMA was clearly lower than in anaerobic conditions in both the inlet and the outlet zones ($p < 0.005$). However, differences for SMA in anoxic conditions between the inlet and outlet zones as well as among the three units were not statistically significant ($p > 0.05$). On average, the SMA reached 22.0 % (HF1), 42.2 (HF2) and 28.4 % (HFc) in anoxic conditions compared to anaerobic conditions. These values indicated the potential reduction of methane production in CWs with recirculation of nitrified effluent (Gonzalo et al., 2017).

Although SMA values are scarce in the literature, the values obtained for the inlet zone were higher than previous reported values for non-aerated HF CWs. Torrijos et al. (2018) reported SMA values of 0.16 and 0.13 mg CH_4-C/kgTS·h for un-planted HF in series at lab scale. Carballeira et al. (2017) reported lower values of 0.008 to 0.013 mg CH_4-C/kgTS·h for planted HF, while López et al. (2015) obtained values of 0.03 to 0.09 mg CH_4-C/kgTS·h. In the present study, the SMA decreased from the inlet zone to the outlet zone, in which values similar to those of the literature were found. Values for SMA in anoxic conditions were not found in the literature, thus comparison is not possible.

Table 6 shows the mean values of methane emission rates from the two measuring campaigns, together with their comparison to potential methane emission derived from SMA values. Higher methane emission rates were measured in the non-aerated unit, which were in the range of those emissions reported in other studies for subsurface horizontal constructed wetlands (Corbella and Puigagut, 2015; de la Varga et al., 2015; Carballeira et al., 2017). However, in spite of similar SMA values in the three units (Table 5), methane emission rates were lower in the aerated units. On average, methane emissions from HF1 were 12 times lower than from HFc. The result agrees with the general trend that a more oxygenated wetland media shows lower methane emission (Mander et al., 2014; Torrijos et al., 2018). Within aerated wetlands, methane emissions from HF1 were lower than from HF2 by a mean factor of 3.2. This could be explained because of the lower VLR in the deeper bed and the operation under non-overloaded conditions, as previously discussed in preceding sections. The emission factor was in the same proportion for the aerated units, i.e., approximately 3 times lower for HF1 than for HF2 unit. However, a greater difference in EF between aerated a non-aerated units was found, as indicated by an EF 56 times lower in HF1 respect to HFc. On the other hand, higher methane emission rates in the inlet zone than in the outlet zone (Table 6) agree with the spatial variation found for SMA (Table 5).

Table 6 presents measured SER of methane in the aerated wetland units, suggesting that methane emissions was only a low percentage of the potential methane generation (i.e. considering the obtained values of SMA). SER of methane was <4 % of PME in anaerobic conditions and still <8 % of anoxic PME on average in aerated units. However, the SER of methane reached high percentages of PME in the non-aerated unit, ranging on average from 18 % PME in anaerobic conditions to 65 % PME in anoxic conditions. These results indicate that aeration effectively

Table 5
Specific methanogenic activity (SMA) measured in anaerobic and anoxic conditions.

Position		SMA (mg CH_4-C/kg TS·h)[a]			Anoxic SMA (mg CH_4-C/kg TS·h)		
		HF1	HF2	HFc	HF1	HF2	HFc
Inlet	Right	0.598 ± 0.000	0.763 ± 0.032	0.449 ± 0.017	0.111 ± 0.017	0.310 ± 0.047	0.131 ± 0.017
	Left	0.522 ± 0.071	0.897 ± 0.141	0.444 ± 0.025	0.128 ± 0.017	0.427 ± 0.031	0.102 ± 0.014
Outlet	Right	0.006 ± 0.000	0.064 ± 0.005	0.136 ± 0.016	0.005 ± 0.000	0.004 ± 0.003	0.038 ± 0.004
	Left	0.006 ± 0.006	0.045 ± 0.006	0.193 ± 0.023	0.005 ± 0.000	0.005 ± 0.003	0.076 ± 0.002
Mean		0.283 ± 0.321	0.442 ± 0.451	0.305 ± 0.164	0.062 ± 0.066	0.187 ± 0.215	0.087 ± 0.040

[a] SMA corresponds to assays carried out with gravel bed samples obtained at the end of the experimentation (campaign C7).

Table 6
Methane surface emission rate (SER) and emission factor (EF) for each wetland unit.

Position	HF1	HF2	HFc
SER (mg CH_4/m^2·d)[a]			
Inlet	128 ± 32	388 ± 256	921 ± 362
Outlet	4 ± 4	29 ± 31	670 ± 145
Mean	66 ± 14	208 ± 144	796 ± 109
EF (%)[a]	0.14 ± 0.01	0.39 ± 0.18	7.78 ± 5.12
SER (% Anaerobic PME)[a]			
Inlet	1.0	3.3	14.6
Outlet	2.9	3.8	28.8
Mean	1.0	3.3	18.4
SER (% Anoxic PME)			
Inlet	4.7	7.4	55.8
Outlet	3.4	43.5	83.5
Mean	4.7	7.9	64.9

[a] SER and EF are the mean of the emissions measured in two campaigns carried out in May and September during periods C3 and C7, respectively. PME: potential methane emission (mg CH_4/m^2·d).

reduced methane emissions but do not inhibit the growth and survival of methanogenic microorganisms.

Registered CO_2 emissions were 5967 ± 4276 mg/m^2·d HFc and HF2 showing higher emission rates than the HF1 unit. However, CO_2 emissions were considered biogenic and do not constitute a contribution to global warming (Dalia et al., 2019). N_2O emission rate was always below the detection limit of the used method.

4. Discussion

Rous et al. (2019) analyzed the relationship between the different design parameters of CWs, including water depth, and their possible influence on aeration efficiency. However, they found no correlation between aeration efficiency and parameters such as water depth or hydraulic loading rate. We must bear in mind that in each of the investigations included in his database ($n = 21$), only experiments with a single bed depth were conducted (Rous et al., 2019). Being in the range of 0.15 to 1.70 m, the average depth in the studies reviewed by Rous et al. (2019) was 0.58 ± 0.34 m and the median was 0.60 m. In the recent review carried out for our study (Section 2.6), again all the research reviewed made use of a single water depth. The only exception is the column aeration efficiency study by Freeman et al. (2018), who, however, only applied a bed depth of 1.4 m in their CW pilot plant. Therefore, the present study is the first investigation in which two HF units with different bed depths are compared, by parallel operation, applying the same influent and the same design of the aeration system and HF units. In addition, these two units were also compared by side-by-side operation with a similar non-aerated unit.

In the present study, we compared the performance of an aerated system with a water depth of 0.8 m (HF1) to two other systems with a water depth of 0.5 m, one aerated in the same conditions (HF2) and the other non-aerated as a control (HFc). In the current study, the gravel size was 12–16 mm and the aeration rate was 0.69 m^3/m^2·h. Vera-Puerto et al. (2022) found that the system with a similar size of coarse gravel (12.7–19.1 mm) to our study, had higher oxygen transfer efficiencies than the system with finer gravel (6.4–12.7 mm) confirming the good selection of the gravel size. Our aeration rate was lower than the range of 2.1–10.7 m^3/m^2·h proposed by Vera-Puerto et al. (2022), in which the lower limit corresponded to the German guidelines for actively aerated filters, but higher than that used by John et al. (2020) (which was in the range of 0.008–0.06 m^3/m^2·h). Therefore, the aeration rate applied in the current study is on the broad range applied by other aerated CW studies.

The observed performance was different for the removal of organic matter than for the removal of ammonia and TN. Stable and high organic matter removal (above 90 % COD removal) was obtained for SLRs up to 80, 40 and 20 g BOD_5/m^2·d for HF1, HF2 and HFc systems, respectively. Thus, introducing aeration but maintaining water depth increased feasible SLR by a factor of 2, while introducing aeration and increasing water depth by 60 % (from 0.5 to 0.8 m) feasible SLR increased by at least a factor of 4. However, VLR controls efficiency better than the SLR, and the aerated units approximately double the feasible VLR than conventional HF for efficient operation (up to 200 and 100 g BOD_5/m^3•d, respectively).

Regarding ammonium removal, the behavior was different, since for a similar efficiency in the two aerated units, HF1 allows a 25 % higher ammonium SLR than HF2, while the VLR was 50 % higher in HF2. The control unit always showed lower ammonia removal efficiency than the aerated units. For similar ammonia removals, the aerated units removed SLR at least 5 times higher loading than the non-aerated unit.

Regarding the mode of aeration, continuous or discontinuous, Rous et al. (2019) found higher values of aeration efficiency for systems with intermittent aeration, although at a statistically non-significant level. The authors consider that there is evidence that intermittent aeration can be more energy efficient while maintaining pollutant removal efficiency. However, John et al. (2020) reported good performance results, including organic matter, ammonia, and total nitrogen removal, of a vertical up-flow saturated CW operated under continuous aeration using reduced air-to-wastewater ratios ranging from 0.5:1 to 4:1. In the latter case, the CWs system contained three units in series, only the second of them being aerated (John et al., 2020). On the other hand, Sossalla et al. (2022) compared continuous aeration in HF systems that was applied along the entire length of the system with other units in which aeration was limited to a fraction of the system, 85 %, 50 % and 35 %. Reducing the aeration zone to 85 % reduced *E. coli* removal, while reductions of up to 50 % can be applied without a large negative impact on conventional water quality parameters. Even, TN removal could be higher in the 50 % aeration zone conditions.

In this sense, only intermittent aeration was applied in the present study, but varying the aeration and non-aeration time, which went from 5 and 3 h to 3 and 5 h, respectively. Under the conditions of this study, the aeration time seems to have no effect on the removal of organic matter, which on the contrary appeared to be controlled by the SLR and the VLR. However, the effect on ammonia removal was clear. Higher ammonia removal rates (>99 % in HF1, >89 % in HF2) were achieved during the 5 h/3 h on/off aeration mode, while during the 3 h/5 h on/off aeration mode, both units showed lower nitrification efficiency, which decreased as the loading rate increased. Virtually, complete ammonia removal could be achieved if the aeration period was conveniently extended.

On the other hand, TN removal varied greatly from 10 to 80 % depending on the operating conditions. However, TN removal showed little difference between units, but was favored by effluent recirculation probably due to the addition of nitrified effluent could find ideal denitrification conditions and sufficient carbon to fuel the process. Because long aeration times were required for ammonia nitrification, it could be concluded that recirculation is still a good option to improve TN removal in aerated CWs. Alternatively, the spatial distribution of aeration could also favor the removal of TN (Sossalla et al., 2022).

Aeration efficiency in biological wastewater treatment systems is expected to be positively related to bed water depth (Freeman et al., 2018; Rous et al., 2019). Rous et al. (2019) calculated the active aeration efficiency from the reviewed articles and concluded that the standard oxygen transfer efficiency was in the range of 1.8–4.1 % per meter of immersion, which is quite similar to the value of 4.7 %/m indicated in Kadlec and Wallace (2009). Higher values ranging 3–16 % per meter can be obtained by diffuse aeration using a disc diffuser, although this is not common practice in CW aeration (Vera-Puerto et al., 2022). The combination of these values with the reduced depths of the water layer in CW results in a very low efficiency of artificial aeration in these systems. Indeed, several authors have reported maximum volumetric oxygen consumption rates of the order of 78–80 g O_2/m^3·d for aerated systems, which would be only slightly higher than the 65 g O_2/m^3·d achieved in non-intermittent aerated vertical flow CWs, although clearly higher than the 27 g O_2/m^3·d that can be achieved in non-aerated horizontal flow CWs (Nivala et al., 2013; Rous et al., 2019). Thus, as highlighted by Rous et al. (2019), citing Kadlec and Wallace (2009), aeration is only justified when its life cycle cost is sufficiently offset by the reduction in capital cost resulting from reducing the size of the wetland. This may only be justified in areas where land for CWs is scarce or at a high price.

Kadlec and Wallace (2009) reported that with aeration, the equivalent wetland size required for BOD_5 removal from domestic wastewater could be reduced by 67 %, while for TSS, the size reduction would be approximately 36 %. The results of the present study indicate that aeration can reduce the area required for CWs with horizontal subsurface flow, in percentages ranging from 50 % to 80 %, depending on the efficiency parameter considered and whether the depth of the bed is increased or not. With half the surface area, aerated HF units can maintain high organic matter removal efficiency and achieve high ammonia removal rates that would not occur in non-aerated HF units. With only 20–25 % of the surface area, an aerated HF unit requires increased water depth (e.g. from 0.5 to 0.8 m) to obtain high organic matter removal efficiencies, but equally low rates and ammonia removal than a non-aerated HF unit. However, it should be mentioned that a similar area reduction with respect to non-aerated HF CWs can be obtained by using intermittently fed passive vertical flow CWs (Rous et al., 2019; Carballeira et al., 2021).

Some other considerations regarding the practical feasibility and sustainability of aerated wetlands should be taken into account. As indicated, reducing the area by introducing artificial aeration generally requires raising the bed and water depth in the CW. CWs generally have depths of 1 m or even less. In addition to economic considerations, the construction of aerated wetlands at greater depths may encounter several difficulties, including problems of structural stability of excavations or various hazards caused by the level and pressure of groundwater on the treatment unit (Freeman et al., 2018). Aeration efficiency must also consider the electrical energy input to blowers, for which less information is often available (Rous et al., 2019). While Rous et al. (2019) did not find a relationship between the airflow and the aeration efficiency, probably due to the variability derived from the fact that these are investigations carried out in different systems and conditions, Freeman et al. (2018) showed that aeration rate significantly controls aeration efficiency, as expected. In the study by these authors, oxygen transfer efficiency decreased significantly from 2.4 % to 1.6 % in 1.5 m deep columns when the aeration rate was increased from 1 L/min to 3 L/min. The same effect was reported by Vera-Puerto et al. (2022). Although increasing the air flow rate significantly increased the oxygen mass transfer coefficient and the specific oxygen transfer rate, the specific oxygen transfer efficiency decreased (Vera-Puerto et al., 2022). Thus, the optimization of the aeration rate is an aspect of great importance in relation to the sustainability of aerated CWs, due to the energy consumption for aeration. In combination with the optimization of the aeration rate, its spatial (Sossalla et al., 2022) or temporal distribution (this study) can help a better energy efficiency in the operation of aerated CWs. Finally, the results of this study also show that the application of higher loading rates in aerated systems did not result in higher solids accumulation, indicating a low risk of clogging. In addition, aeration effectively reduced methane generation limiting greenhouse gas emissions.

5. Conclusions

This study reports the performance comparison of an aerated HF1 system with a water depth of 0.8 m with two other systems with a water depth of 0.5 m, HF2 aerated under the same conditions and HFc not aerated as a control unit. In the aerated units, the SLR ranged from 20 to 80 g $BOD_5/m^2 \cdot d$ and from 3.7 to 6.7 g $TN/m^2 \cdot d$, while it ranges from 6 to 23 g $BOD/m^2 \cdot d$ and from 1.1 to 1.7 g $TN/m^2 \cdot d$ in the control unit.

The main conclusions of the present study are the following:

- In terms of organic matter removal, the control unit and the aerated HF1 showed sufficient capacity to cope with the applied SLR and achieved a high-quality effluent. In contrast, aerated HF2 (shallower bed depth than HF1) showed more limited capacity. The best COD removal, >90 %, was achieved for an SLR up to 80 g $BOD_5/m^2 \cdot d$ for HF1, but for <40 g $BOD_5/m^2 \cdot d$ for HF2 and 20 g $BOD_5/m^2 \cdot d$ for HFc.
- A longer aeration period of 5 h instead of 3 h in each 8 h cycle favored advanced ammonia removal, which reached mean effluent concentrations of 0.1 ± 0.2 and 1.2 ± 2.5 mg NH_4^+- N/L for HF1 and HF2 units, respectively. TN removal was favored by effluent recirculation in the aerated units, leading to 50–80 % TN removal.
- With artificial aeration, the SLR could be increased by a factor of 4 if the bed (and water) depth increased from 0.5 m to 0.8 m, and only by a factor of 2 if the depth remained the same. Thus, aeration can reduce the surface needed for CWs with horizontal subsurface flow, in percentages ranging between 50 % and 80 %.
- The specific nitrifying and denitrifying activities of the accumulated biomass increased greatly in HF2 relative to HFc (almost 4-fold), but mildly in HF1 compared to HFc (almost 2-fold). Specific activities and biomass content were higher in the inlet part of the aerated wetlands, while the non-aerated unit showed more uniform values. The risk of clogging was assessed to be low.
- The SMA was clearly lower in anoxic conditions than in anaerobic conditions in both the inlet and the outlet zones of the CW, what indicates the potential reduction of methane production in CWs with recirculation of nitrified effluent.
- The aeration effectively reduced methane emission in both aerated units (in HF1 12 times lower than HFc). The lower methane emissions of the deeper unit (in HF1 3 times lower than HF2) was due to the lower VLR applied and operation under non-overload conditions.

CRediT authorship contribution statement

Ana Pascual: Investigation, Methodology, Validation, Formal Analysis, Original draft preparation.
Juan A. Álvarez: Conceptualization, Resources, Methodology, Supervision, Writing-reviewing.
Manuel Soto: Conceptualization, Methodology, Supervision, Data analysis, Software, Data discussion, Writing-reviewing.
Carlos A. Arias: Conceptualization, Data discussion, Supervision, Writing-reviewing.
David de la Varga: Conceptualization, Methodology, Formal Analysis, Original draft preparation, Writing-reviewing.
Rene Killian: Conceptualization, Writing-reviewing.
Dion Van Oirschot: Conceptualization, Writing-reviewing.

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.

Data availability

Data will be made available on request.

Acknowledgements

This work was supported by the EU's seventh framework program for research, technological development and demonstration under grant agreement n° 605445 (HIGHWET project).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.168257.

References

Aguilar, L., Gallegos, A., Pérez, L.M., Arias, C.A., Rubio, R., Haulani, L., García, J., Pallarés, M., de Pablo, J., Morató, J., 2021. Effect of intermittent induced aeration on nitrogen removal and denitrifying-bacterial community structure in Cork and gravel vertical flow pilot-scale treatment wetlands. J. Environ. Sci. Health A 56 (10), 1121–1130. https://doi.org/10.1080/10934529.2021.1967652.
Aguilar, L., Pérez, L.M., Gallegos, A., Fores, E., Arias, C.A., Bosch, C., Verdum, M., Jove, P., de Pablo, J., Morató, J., 2022. Effect of aeration on nitrogen removal-associated microbial community in an innovative vertical cork-based constructed wetland for winery wastewater treatment. Ecol. Eng. 185, 106781 https://doi.org/10.1016/j.ecoleng.2022.106781.
Álvarez, J.A., Zapico, C.A., Gómez, M., Presas, J., Soto, M., 2003. Anaerobic hydrolysis of a municipal wastewater in a pilot-scale digester. Water Sci. Technol. 47 (12), 223–230 (PMID: 12926692).
APHA, 2005. Standard Methods for the Examination of Water and Wastewater, 21st ed. American Public Health Association/American Water Works Association/Water Environment Federation, Washington, DC.
Brix, H., 1989. Gas exchange through dead culms of reed, phragmites australis (cav.) trin. ex steudel. Aquat. Bot. 35, 81–98.

Carballeira, T., Ruiz, I., Soto, M., 2017. Methanogenic activity of accumulated solids and gas emissions from planted and unplanted shallow horizontal subsurface flow constructed wetlands. Ecol. Eng. 98, 297–306. https://doi.org/10.1016/j.ecoleng.2016.10.079.

Carballeira, T., Ruiz, I., Soto, M., 2021. Improving the performance of vertical flow constructed wetlands by modifying the filtering media structure. Environ. Sci. Pollut. Res. 2021, 1–13. https://doi.org/10.1007/S11356-021-14389-1.

Castellar, J.A., 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. Clean. Prod. 340, 130660 https://doi.org/10.1016/j.jclepro.2022.130660.

Corbella, C., Puigagut, J., 2015. Effect of primary treatment and organic loading on methane emissions from horizontal subsurface flow constructed wetlands treating urban wastewater. Ecol. Eng. 80, 79–84. https://doi.org/10.1016/j.ecoleng.2014.09.071.

Dalia, J., Antunes, M., Almeida, S., 2019. Life cycle assessment and costing of wastewater treatment systems coupled to constructed wetlands. Resour. Conserv. Recycl. https://doi.org/10.1016/j.resconrec.2019.04.034.

Dotro, G., Langergraber, G., Molle, P., Nivala, J., Puigagut, J., Stein, O.R., von Sperling, M., 2017. Treatment Wetlands. (Biological Wastewater Treatment Series, Volume 7). IWA Publishing, London, UK. https://doi.org/10.2166/9781780408774, 172p. eISBN: 9781780408774; available at:. (accessed 30 April 2019).

Freeman, A.I., Surridge, B.W.J., Matthews, M., Stewart, M., Haygarth, P.M., 2018. New approaches to enhance pollutant removal in artificially aerated wastewater treatment systems. Sci. Total Environ. 627, 1182–1194.

Gonzalo, O.G., Ruiz, I., Soto, M., 2017. Integrating pretreatment and denitrification in constructed wetland systems. Sci. Total Environ. 584–585 (2017), 1300–1309. https://doi.org/10.1016/j.scitotenv.2017.01.217.

Green Infrastructure Guide, 2014. United Nations Environment Programme (UNEP). Available at: https://www.unep.org/resources/publication/green-infrastructure-guide-water-management (accessed 22 July 2023).

Headley, T., Nivala, J., Kassa, K., Olsson, L., Wallace, S., Brix, H., van Afferden, M., Müller, R.A., 2013. *Escherichia coli* removal and internal dynamics in subsurface flow ecotechnologies: effects of design and plants. Ecol. Eng. 61, 564–574. https://doi.org/10.1016/j.ecoleng.2013.07.062.

John, Y., Langergraber, G., Adyel, T.M., Jr, Emery David, V., 2020. Aeration intensity simulation in a saturated vertical up-flow constructed wetland. Sci. Total Environ. 708, 134793 https://doi.org/10.1016/j.scitotenv.2019.134793.

Kadlec, R., Wallace, S., 2009. Treatment Wetlands, 2nd ed. CRC Press, Boca Raton, FL.

López, D., Fuenzalida, D., Vera, I., Rojas, K., Vidal, G., 2015. Relationship between the removal of organic matter and the production of methane in subsurface flow constructed wetlands designed for wastewater treatment. Ecol. Eng. 83, 296–304. https://doi.org/10.1016/j.ecoleng.2015.06.037.

Lv, T., Wu, S., Hong, H., Chen, L., Dong, R., 2013. Dynamics of nitrobenzene degradation and interactions with nitrogen transformations in laboratory-scale constructed wetlands. Bioresour. Technol. 133, 529–536. https://doi.org/10.1016/j.biortech.2013.02.003.

Mander, Ü., Dotro, G., Ebie, Y., Towprayoon, S., Chiemchaisri, C., Nogueira, S.F., Jamsranjav, B., Kasak, K., Truu, J., Tournebize, J., Mitsch, W.J., 2014. Greenhouse gas emission in constructed wetlands for wastewater treatment: a review. Ecol. Eng. 66, 19–35. https://doi.org/10.1016/j.ecoleng.2013.12.006.

Murphy, C., Rajabzadeh, A.R., Weber, K.P., Nivala, J., Wallace, S.D., Cooper, D.J., 2016. Nitrification cessation and recovery in an aerated saturated vertical subsurface flow treatment wetland: field studies and microscale biofilm modelling. Bioresour. Technol. 132, 125–132. https://doi.org/10.1016/j.biortech.2016.02.065.

Navidi, W., 2006. Statistics for Engineers and Scientists. McGraw-Hill, México.

Nivala, J., Wallace, S., Headley, T., Kassa, K., Brix, H., van Afferden, M., Müller, R., 2013. Oxygen transfer and consumption in subsurface flow treatment wetlands. Ecol. Eng. 61, 544–554. https://doi.org/10.1016/j.ecoleng.2012.08.028.

Nivala, J., Boog, J., Headley, T., Aubron, T., Wallace, S., Brix, H., Mothes, S., van Afferden, M., Müller, Roland A., 2018. Side-by-side comparison of 15 pilot-scale conventional and intensified subsurface flow wetlands for treatment of domestic wastewater. Sci. Total Environ. 658, 1500–1513. https://doi.org/10.1016/j.scitotenv.2018.12.165.

Pascual, A., de la Varga, D., Arias, C.A., Van Oirschot, D., Kilian, R., Álvarez, J.A., Soto, M., 2016. Hydrolytic anaerobic reactor and aerated constructed wetland systems for municipal wastewater treatment - HIGHWET project. Environ. Technol. Jan 38 (2), 209–219. https://doi.org/10.1080/09593330.2016.1188995.

Redmond, E., 2012. Nitrogen removal from wastewater by an aerated subsurface flow constructed wetland. Master thesis. University of Iowa (US). https://doi.org/10.17077/etd.jd4ll2e7.

Rous, V., Vymazal, J., Hnátková, T., 2019. Treatment wetlands aeration efficiency: a review. Ecol. Eng. 136, 62–67. https://doi.org/10.1016/j.ecoleng.2019.06.006.

Suhad, A.A.A.N.Almuktar, Abed, Suhail N., Scholz, Miklas, 2018. Wetlands for wastewater treatment and subsequent recycling of treated effluent: a review. Environ. Sci. Pollut. Res. 25, 23595–23623. https://doi.org/10.1007/s11356-018-2629-3.

Sossalla, N.A., Nivala, J., Escher, B.I., Schlichting, R., van Afferden, M., Müller, R., Reemtsma, T., 2022. Impact of various aeration strategies on the removal of micropollutants and biological effects in aerated horizontal flow treatment wetlands. Sci. Total Environ. 828, 154423 https://doi.org/10.1016/j.scitotenv.2022.154423.

Sowińska-Świerkosz, B., García, J., 2022. What are nature-based solutions (NBS) setting core ideas for concept clarification. Nature-Based Solutions 2, 100027. https://doi.org/10.1016/j.nbsj.2022.100009.

Stefanakis, A., 2019. The role of constructed wetlands as green infrastructure for sustainable urban water management. Sustainability 11 (24), 6981. https://doi.org/10.3390/su11246981.

Štrbac, S., Kašanin-Grubin, M., Pezo, L., Stoji'c, N., Loncar, B., Curcic, L., Pucarevi'c, M., 2023. Green infrastructure designed through nature-based solutions for sustainable urban development. Int. J. Environ. Res. Public Health 20, 1102. https://doi.org/10.3390/ijerph20021102.

Torrijos, V., Ruiz, I., Soto, M., 2018. Microbial activities and process rates in two-step vertical and horizontal subsurface flow gravel and sand filters. Water Air Soil Pollut. 229, 290. https://doi.org/10.1007/s11270-018-3934-7.

de la Varga, D., Ruiz, I., Díaz, M.A., Soto, M., 2013. Avoiding clogging in constructed wetlands by using anaerobic digesters as pre-treatment. Ecol. Eng. 52, 262–269.

de la Varga, D., Ruiz, I., Álvarez, J.A., Soto, M., 2015. Methane and carbon dioxide emissions from constructed wetlands receiving anaerobically pretreated sewage. Sci. Total Environ. 538, 824–833.

Vera-Puerto, I., Campal, J., Martínez, S., Cortés-Rico, L., Coy, H., Tan, S., Arias, C.A., Baquero-Rodríguez, G., Rosso, D., 2022. Effects of environmental conditions and bed configuration on oxygen transfer efficiency in aerated constructed wetlands. Water 2022 (14), 3284. https://doi.org/10.3390/w14203284.

Vymazal, J., 2010. Constructed wetlands for wastewater treatment. Water 2, 530–549. https://doi.org/10.3390/w2030530.

Vymazal, J., Kröpfelová, L., 2008. Wastewater Treatment in Constructed Wetlands with Horizontal Sub-Surface Flow. In: Environmental Pollution (EPOL 14). Springer, Dordrecht. https://doi.org/10.1007/978-1-4020-8580-2.

Wallace, S., Nivala, J., Meyers, T., 2008. Statistical analysis of treatment performance in aerated and nonaerated subsurface flow constructed wetlands. In: Vymazal, J. (Ed.), Wastewater Treatment, Plant Dynamics and Management in Constructed and Natural Wetlands. Springer, Dordrecht. https://doi.org/10.1007/978-1-4020-8235-1_15.

Wallace, S., Oirschot, D., Stefanakis, A., 2020. Wetland Technology: Practical Information on the Design and Application of Treatment Wetlands - Chapter 5.7. IWA Publishing, pp. 105–107.

Zhang, J., Wu, H., Hu, Z., Liang, S., Fan, J., 2014. Examination of oxygen release from plants in constructed wetlands in different stages of wetland plant life cycle. Environ. Sci. Pollut. Res. 21, 9709–9716. https://doi.org/10.1007/s11356-014-2905-9.

Annex IV

Ecological Engineering 169 (2021) 106311

Contents lists available at ScienceDirect

Ecological Engineering

journal homepage: www.elsevier.com/locate/ecoleng

Nature based solutions for winery wastewater valorisation

Ana Pascual [a,*], Rocio Pena [a], Santiago Gómez-Cuervo [a], David de la Varga [b], Juan A. Alvarez [a], Manuel Soto [b], Carlos A. Arias [c,d]

[a] Department of Environmental Technologies, AIMEN Technology Centre, Pontevedra, Spain
[b] Department of Chemistry, University of A Coruña, Spain
[c] Department of Biology - Aquatic Biology, Aarhus University, Ole Worms Allé 1, 8000 Aarhus C, Denmark
[d] WATEC Aarhus University Centre for Water Technology, NyMunkegade, Bldg. 1521, 8000 Aarhus C, Denmark

ARTICLE INFO

Keywords:
Constructed wetlands
Hybrid systems
Anaerobic reactor
Wastewater
Winery

ABSTRACT

To valorise winery treated wastewater and to produce suitable water for the irrigation of vineyards, or other factory operations, the WETWINE project set up a combination of a hydrolytic up-flow sludge blanket (HUSB) reactor and constructed wetlands (CW). The WETWINE plant is located in Tomiño (Pontevedra, Spain) and is being validated at the demo-scale at Santiago Ruiz Winery (NW Spain). The plant consisted of a HUSB reactor for removing suspended solids and hydrolyse organic matter, followed by two vertical subsurface flow constructed wetland (VF) operating in parallel and to provide further treatment, a horizontal subsurface flow constructed wetland (HF) operating in series. After two years of continuous monitoring and regardless in the variation of flow and load, the hybrid system achieved average removal efficiencies of over 93% for total suspended solids and chemical oxygen demand, receiving an overall surface loading of up to 115 g COD/m^2·d. Total nitrogen removal reached 62% with an average concertation of 21 mg N/ L. The WETWINE plant was also able to increased the pH in the effluent from 5.5 at the influent to 7.2.

1. Introduction

Worldwide, the wine industry produces large quantities of wastewater as a result of the different operations demanded by the processes: namely harvesting, pressing, the fermentation phases and the bottling of the wine (Mulidzi, 2007). The resultant wastewater constitutes a serious environmental for the wine producing countries due to the high organic loads (COD, 300 to 49,000 mg/L), the highly fluctuating concentrations of total suspended solids (TSS, from 12 to 18,000 mg/L) and acidic characteristics of the wastewater (pH = 3–5) (Masi et al., 2015).

Conventional treatment systems are not effective for the treatment of winery wastewater due to the fact that wineries show highly variable discharge flows and loadings, both daily as well as seasonally (Serrano et al., 2011; De la Varga, 2014). During the course of each year, wastewater flow and the concentration of the wastewater produced, vary in relation to the working period (vintage, racking or bottling), the size of the winery, the product (red, white and specialty vines, and subproducts such as spirits) or the strategy applied for the management of waste (De la Varga, 2014; Masi et al., 2015). In addition, these systems have high operating costs, demand extensive maintenance if they are to adapt to wastewater continues characteristics changes typical of the wine industry.

Constructed wetlands (CWs) are engineered wastewater treatment systems, designed and optimized to mimic the processes that occur in natural wetlands (Vymazal, 2014). The granular media, vegetation, soils, and their associated microbial assemblages are combined to treat wastewater effectively (Pascual et al., 2016). CW constitutes a flexible and robust technology that can adapt to very different organic and hydraulic loadings and has shown promising results when treating winery effluent (Uggetti et al., 2011; De la Varga et al., 2013a, 2013b; De la Varga et al., 2016; Brix, 2017). To achieve better results, CW configurations can be combined (hybrid systems), to enhance pollutant removal efficiency, especially nitrogen removal (Vymazal, 2009) as well to tackle highly strength wastewaters (Masi et al., 2015; Torrijos et al., 2016).

CW systems have been used to treat industrial effluents from petrochemical, dairy, meat processing, abattoir, and pulp and paper factory production (Vymazal, 2014). Brewery, winery, tannery and olive mills wastewaters have been recently added to CW applications Vymazal (2013) reported the use of CWs for the treatment of industrial wastewater with influent concentrations ranging up 10,000–24,000 mg COD/

* Corresponding author.
E-mail addresses: apascual@aimen.es (A. Pascual), m.soto@udc.es (M. Soto), carlos.arias@bio.au.dk (C.A. Arias).

https://doi.org/10.1016/j.ecoleng.2021.106311
Received 27 February 2020; Received in revised form 28 May 2021; Accepted 9 June 2021

L and up to 496 mg NH_4^+-N/L. However, there are no general rules for selecting the most suitable type of CW for certain industrial wastewater or even to treat urban wastewater. Every wastewater is specific the decision of which technology or CW to be used must be studied according to several conditions, among others: type of wastewater, land availability, flow and pollutant load, effluent discharge limits. (De la Varga et al., 2016).

The most common CWs used for treating polluted waters are: free water surface systems, horizontal subsurface flow systems (HF) and vertical subsurface flow systems (VF). To improve performance by enhancing the removal of pollutants and nutrients, a combination of different wetlands types can be used, e.g. the so called hybrid systems, which can combine HF and VFs HF are usually operated in conditions of permanent water saturation which limits oxygen transfer and therefore nitrification only occurs at a low rate. Pulsed fed VF operate mostly unsaturated and provide good conditions for nitrification, but denitrification in these systems is limited (Brix and Arias, 2005). Therefore, in hybrid systems, the strengths and weaknesses of each type of system are combined and in consequence, it is possible to produce an effluent low concentrations of organic matter as well as effective total nitrogen removal (Vymazal, 2009). Hybrid systems are flexible and selecting the order of the treatment train will depend on the pollutants to treat and the discharge targets. The placement of the systems is flexible and can even allow the recirculation of flows among the structures or to the primary treatment to enhance performance of pollutant removal (Brix and Arias, 2005).

Additionally, clogging prevention and phosphorus removal along time are issues that have not yet been completely solved when CWs are established. CW media clogging can occur when the systems are not properly design or operated. Although CW clogging dynamics are extremely complex and still not well-understood, suspended solids as well as organic loads influent concentration are a determinant factors when clogging occurs. The use of anaerobic digesters as pre-treatment can achieve high suspended solids removal, organic matter removal and therefore reduce wetland clogging risks (Álvarez et al., 2008; Ruiz et al., 2008; Ruiz et al., 2010; Pedescoll et al., 2011). Furthermore, the combination of both technologies, namely CWs and anaerobic digesters can be of great interest when combined as both are natural based solutions implying low cost, sustainability, simplicity of construction, operation and maintenance.

Excess sludge generated in anaerobic digesters may be treated in sludge treatment wetlands (STW). STWs is a type of CW consisting of several parallel beds that are fed sequentially at loadings that depend on the characteristics of the sludge, the local climatic conditions and the size of the beds. The planted beds are filled with successive layers of gravel and soil/sand materials that favours the establishment (De la Varga et al., 2016). In STWs the sludge is pumped intermittently into the different beds, alternating dosing regimens and resting periods. As sludge builds up, the rhizomes will develop and penetrate into the sludge layer and as the reeds grow, dewatering of the sludge happens by the combination of evapotranspiration by the plants and the filtering effect of the bed. Additionally to the stabilization and dewatering, Uggetti et al. (2012) reported nitrogen and phosphorus reductions due to sludge mineralization, ammonification and plant uptake.

The aim of the WETWINE project (SOE1/P5/E0300) is to establish a system treat water and the sludge produced by the primary treatment and to recover resources from the of wine industry, using CWs, a low-cost natural based technology that can be established by the wine producers in the SUDOE regional area (Portugal, Spain and southern France). Since the region is limited in water resources, is of outmost importance to reclaim water suitable for agricultural irrigation that meets the new Regulation, 2020/741 or and comply with Spanish National Regulation RD 1620/2007. For this purpose, a demonstration plant was established in northwest of Spain (Tomiño, Pontevedra) with the combination of an anaerobic reactor (HUSB) as primary treatment, CWs for the treatment of the waters and a CWs for the dewatering and mineralization of the sludge produced by the primary treatment. This work reports two years results from wastewater treatment line of the WETWINE plant.

2. Materials and methods

2.1. WETWINE pilot plant

The WETWINE pilot plant was built in the facilities of the Santiago Ruiz Winery in Tomiño (Spain, 41°59′33.9"N 8°43′15.0"W). The winery has 35 ha of planted vineyard, it produces 368.000 L/y of wine resulting in large volumes of highly loaded wastewater. During the two months of the grapes harvest, the estimate water produced is 620 m^3 while during the one month grape fermentation period the wastewater produced reaches 130 m^3 for the remaining 9 months of off-peak period, the volume of wastewater and the pollutant load decreases considerably to a total of 648 m^3.

The winery effluents are constituted by soluble and insoluble phases. The soluble phase consists of organic compounds, such as polyphenols, organic acids, polysaccharides, lignin, reduced sugars, and melanoidins from the grapes, oenological deposits and cleaning products. The insoluble phase is formed by particles of various sizes such as scrapes, skins, grapes, seeds, plant debris, tartrate crystals, etc. (De la Varga, 2014). Because the insoluble compounds may be present, a pre-treatment step was considered necessary to remove them from the influent.

The WETWINE pilot plant consisted of two treatment lines: wastewater treatment line and sludge treatment line. The wastewater treatment line of the WETWINE pilot plant consisted of a Hydrolytic Up-Flow Sludge Blanket (HUSB) anaerobic reactor followed by two parallel unsaturated vertical flow constructed wetlands, VF, and a horizontal flow constructed wetland, HF. The system was completed with four sludge treatment wetlands that received the sludge generated by the HUSB reactor. At the end of sludge treatment wetlands, an adsorption filter was placed well to remove nutrients. Fig. 1 presents the diagram of the pilot plant design.

The plant was fitted with the necessary pumps and wells to ensure effective water flow and possibility for sampling. The influent wastewater flowed by gravity to 40 m^3 underground storage tank. From the storage tank a 0.5 kW sewage pump feeds the HUSB digester. The pumping systems was installed with all the required structures: a control valve and a non-return valve as well as the necessary safety features Flow was measured with a magnetic flowmeter to record instantaneous flow as well as accumulated flow. The pump and the flow meter, allowed accurately control of the wastewater pumped to the pilot plant and to calculate the actual loadings. Raw influent wastewater was pumped to the HUSB at an average flow rate of 1275 ± 1181 L/d. The effluent form the HUSB was conducted to a 1 m^3 pumping well (P1) fitted with a peristaltic pump to pump the wastewater to the VF. The feeding to the two VF beds was done alternatively, between 2 and 4 pulses per day. Following this stage, the wastewater was conducted to a saturated HF of effective depth of 1.1 m and water level 5 mm below, to avoid foul odours and the proliferation of potential vectors. After the HF the water was conducted to a final 5 m^3 tank. The tank stores the treated water to be used for irrigation only if the quality meets the irrigation standards, on the contrary if the quality is not appropriate for irrigation, is discharged to the municipal sewer for further treatment. To reuse the water for irrigation the new national Regulation (2020/741) is already being enforced. The new rules will apply from 26 June 2023 and are expected to stimulate and facilitate water reuse in agriculture in the EU. Moreover, this final tank is also fitted with a 0.5 kW pump to recirculate the water to storage tank if necessary.

2.2. HUSB reactor

The primary of the WETWINE pilot plant uses a HUSB reactor. The

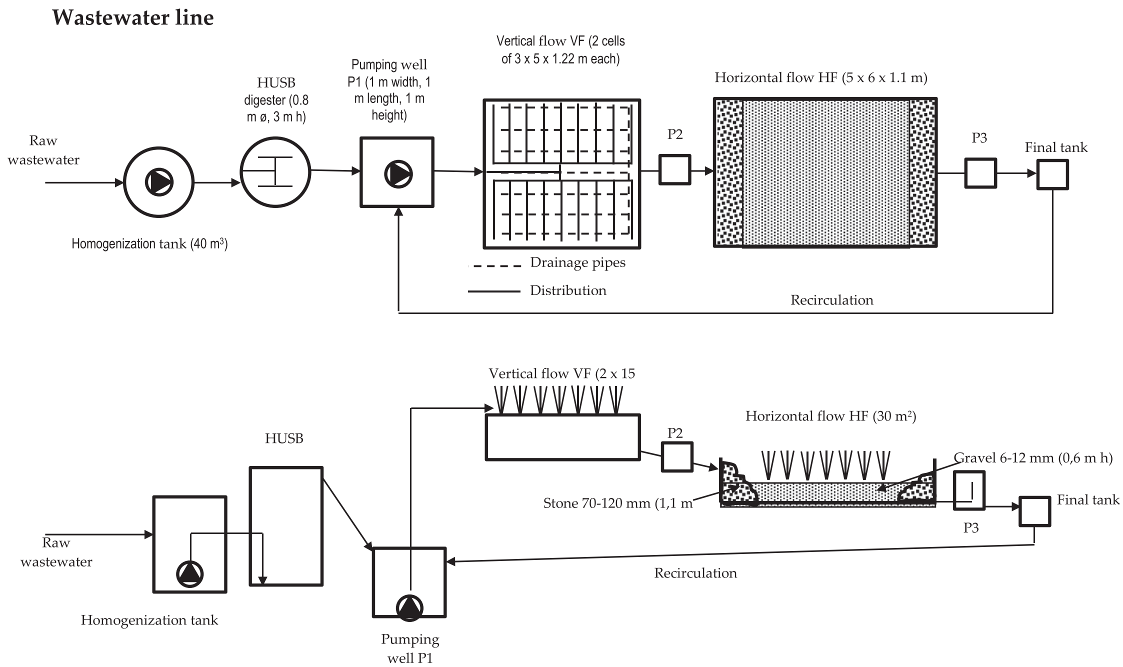


Fig. 1. Configuration wastewater treatment line of WETWINE plant (top view and profile).

latter consisted 3 m high Ø 0.8 m diameter hermetic cylindrical tank and with an effective depth of 2.5 m, for a total active volume of 1.51 m^3. The design hydraulic retention time (HRT) during harvest was 11 h (minimum HRT). For an optimum solids retention, the up-flow velocity of water in this HUSB should not exceed 0.3 m/h (at most 1 m/h) In addition, starting from the bottom, this HUSB has four sample ports to control the sludge volume accumulated in the reactor. In this kind of digester, methane production is avoided or minimized (Álvarez et al., 2003), and should only working the hydrolytic phase, then the gas/liquid/solid separator are not needed, therefore, a HUSB is not equipped with a gas/liquid/solid separator. The organic loading rate (VLR) calculated in the range of 2–6 g COD/L·d (Álvarez and Soto, 2011).

2.3. CWs units

The water treatment line consisted of two constructed wetland steps in series. The first step consisted of two 15 m^2 unsaturated VF working in parallel that are alternatively fed. The bottom of of the beds and the walls were lined with a HDPE 0.5 mm tick geomembrane, to ensure impermeability. The total height of both cells was 1.22 m (1 m for the effective filter depth and 0.22 m freeboard). The wetland was built directly on the ground, confined with marine formwork walls.

The VFs were fitted with a drainage and influent manifolds to evacuate and distribute waters effectively. The drainage manifold was built with of Ø 75 mm pipes perforated pipes () placed at the bottom of the wetland, engulfed with 20–30 mm of diameter (200 mm) gravel to favour drainage, while the influent distribution system consisted of pipes (Ø 40 mm) placed at the surface of the wetland. The distribution system were perforated with 6 mm holes, at 0,5 m intervals. The VF beds were filled with a series of layers of granitic gravel and sand, from bottom to top: 6–12 mm of diameter (0,10 m depth), 2–4 mm of diameter (0,60 m depth) and a sand layer of 1–2 mm (0,10 m depth).

Once the water trickles through the VF, it flows by gravity to a 30 m^2 HF 5 m wide and 6 m long The bed was excavated and lined with 0.5 mm HDPE and protected with a geotextile. The influent distribution system consisted of a PVC pipe (Ø 75 mm) placed at 910 mm from the bottom of the wetland. At the end of the wetland, a 75 mm in diameter effluent pipe was installed, perpendicular to the direction of the flow and at the bottom of the bed. To minimize the risk of clogging and to evenly distribute and collect the water, sections of, 0,5 mm of coarse gravel (Ø 60–80 mm) were placed at the inlet and the outlet sections of the wetland. The bed was filled with Ø 8–12 mm gravel up to 0,6 m high. The water level in the wetland was regulated with a flexible pipe, installed at a well adjacent to the bed.

Common reed (*Phragmites australis*) was the plant selected for the wetlands and planted at a density of 16 plants/m^2 in all the units.

2.4. Sampling and analysis

During the operation, the pumps were periodically calibrated and the flow was adjusted to the desired flow, and the actual flow was measured. Grab samples were collected either once or twice a week in the following points: 1) raw wastewater (inlet HUSB), 2) outlet HUSB (inlet VF), 3) outlet VF (inlet HF), and 4) outlet HF. The samples were transported under refrigeration and analysed in the laboratory for TSS, COD, anions, cations and total nitrogen. Onsite measurements of pH and electrical conductivity (EC) were taken using a calibrated Dual Start pH-meter and a GLP32 Crison EC-meter, respectively. The determination of COD and TSS were done following the Standard Methods (APHA-AWWA-WPCF, 2005). The ions fluoride (F-), chloride (Cl^-), bromide (Br^-), nitrite (NO^{2-}), nitrate (NO^{3-}), phosphate (PO_4^{3-}), sulphate (SO_4^{2-}), lithium (Li^+), sodium (Na^+), potassium (K^+), ammonium (NH_4^+), calcium (Ca^{2+}) and magnesium (Mg^{2+}) were analysed through ion chromatography (Ionic Chromatograph Metrohm, model 882/863 Compact IC Plus.

2.5. Calculations and statistical methods

The WETWINE plant was operated for two years (2017–2019). The start-up of the WETWINE pilot plant was carried out in the beginning of July 2017 by feeding the plant with 500 L/d of diluted wastewater generated from bottling tasks at the winery. Steady state operation lstarted after August 23, 2017 (Period I). Thereafter, the regime of operation of the pilot plant over the study period depended on the seasonal variations of the winery production as well as on the flow storage capacity of the storage tank (Fig. 1a). The storage capacity was used during the first harvesting period to delay the high load operation, but not during the second harvesting period.

For the calculation and analysis of the effect of different parameters on plant performance, the current study was divided into ten main operating periods. The criterion applied was the homogeneity in the COD treatment efficiency of the VF unit, because this step has shown to determine the overall efficiency of the treatment plant.

For the statistics analysis, one-way and two-way analysis of variance (ANOVA) was used to compare the data sets (Miller et al., 2002) and te least-squares fitting (single and multiple linear regression) was and the square of the coefficient R^2 determination, was used to estimate performance parameters, the adjusted R^2, the statistical F-value and probability (p) were also calculated using Microsoft Excel statistical analysis software package.

3. Results and Discussion

3.1. The operation of the WETWINE plant

Table 1 shows the characteristics of the influent and effluent water from the winery during the ten operating periods. In the periods following the start-up (I, II, III and IV), the tasks of the winemaking operation, fermentation, racking, etc., were carried out at the winery normally. In these periods, the wastewater presented high TSS concentrations (e.g., average value of 767 m TSS/L in period III) and COD (e.g., average value of 10,577 mg COD/ L in period III). Regardless of the high concentrations, it was possible to continue treating high loaded water until period V by diverting water to a 40,000 L storage tank. Periods VI and VII (from April to September 2018) correspond to a low wine production period at the factory, so the wastewater generated is relatively lightly loaded (average values of 1192 mg COD/L and 191 mg TSS/L during period VII). The following period VIII corresponded to the second grape harvesting season. Therefore, high-loaded wastewater (average values of 4990 mg COD/L and 497 mg TSS/L in period VIII) was treated the WETWINE plant. The last monitored periods (IX and X) with very were of low activity at the factory, with low load. The average hydraulic, volumetric and surface loading rate for the HUSB and the CWs (HRT, VLR and SLR respectively) are reported in Table 2.

3.2. Performance of the HUSB reactor

Fig. 2 shows the percentage of TSS and COD removal in the HUSB for the different monitored periods. The HUSB effectively removed the TSS during VI, VII, and VIII with over 50% removal in all cases. During II, III, V and IX periods, the average removal percentage of TSS was low, and during I, IV and X periods the removal percentages were very low or even negative suggesting release of TSS. The low TSS removal during periods III, IV and V can be attributed to a high up-flow velocity in the HUSB (4.4 m/h in period IV). This was due to a pumping time of the feeding pump causing a high instantaneous flow rate in the HUSB reactor and resuspension of the solids of the previously retained solids. After period V, the ON/OFF timing of the feeding pump was adjusted to meet the up flow velocity below 1 m/h.

The data shows that the TSS removal (%TSS_r) increased as the concentration of TSS increased at the influent (TSS_{in}). After day 230 (VI period), the retention of TSS (HRT_{TSS}) at the HUSB was higher than 50% whenever the concentration of TSS in the influent exceeded 200 mg/L. Besides, a minimum TSS effluent (TSS_{EF}) concentration of 57 mg/L was found for near zero TSS influent concentrations. From the 230 operation days that the systems was followed the HUSB influent and effluent presented average values of 245 ± 69 mg TSS/L and 104 ± 36 mg TSS/L, respectively, an average TSS removal of 58%. On the other hand, COD average removal in the HUSB was only 10% during the same period. During the VI monitoring period, a positive correlation between the TSS influent concentration and the TSS removal efficiency ($R^2 = 0.92$, $p < 0.01$) was found. No correlation between %TSS removal and HRT or VLR ($R^2 < 0.51$) was found.

Throughout the operating time, the influent presented a pH average value of 5.1 ± 0.6 while the effluent presented values of 5.20 ± 0.54. It can be seen that the HUSB had no significant effect on pH ($p = 0.75$). Furthermore, the influent pH had no effect on the TSS removal ($R^2 = -0.15$, $p = 0.28$). Thus, neutralization of the influent to the HUSB is not recommended as it does not favour TSS removal and maintaining a low pH constrains methane generation in the anaerobic reactor. Bacteria

Table 1
Characteristics of influent and effluent from each unit of the WETWINE plant.

		pH				EC (μS/cm)				TSS (mg/L)				COD (mg/L)			
Period and days (n[a])		Inf.	HUSB	VF	HF	Inf.	HUSB	VF	HF	Inf.	HUSB	VF	HF	Inf.	HUSB	VF	HF
I) 0–40 (8)	X̄[b]	4.62	4.57	5.97	5.48	711	712	669	759	154	143	137	16	2865	2961	976	1091
	σ[c]	0.79	0.78	1.14	0.96	204	217	164	211	90	106	102	8	1749	1970	777	686
II) 41–100 (9)	X̄	4.81	4.79	7.58	6.27	2667	2648	2094	2168	368	196	90	24	6052	5682	645	1093
	σ	0.53	0.56	0.51	0.90	580	787	673	880	331	74	98	15	1518	1291	370	569
III) 101–150 (3)	X̄	4.99	4.93	5.71	5.11	3147	3073	2500	1827	767	587	200	49	10,577	10,867	4870	4025
	σ	0.78	0.72	1.20	0.54	681	585	356	738	126	61	110	31	3619	4368	2760	3241
IV) 151–186 (4)	X̄	5.45	5.16	7.74	6.85	2494	2901	2899	2609	846	1007	217	27	4097	5824	501	1641
	σ	0.49	0.20	0.43	0.80	935	1118	904	832	1041	785	265	15	2457	2320	260	1256
V) 187–230 (5)	X̄	4.91	4.99	7.07	6.77	1498	1539	975	813	465	317	217	24	6510	6400	264	343
	σ	0.16	0.12	0.27	0.31	348	366	279	349	326	192	259	13	2402	2855	342	280
VI) 231–270 (3)	X̄	5.47	5.54	6.72	7.19	478	640	671	527	316	106	142	29	593	840	372	141
	σ	0.97	0.89	0.05	0.49	33	159	223	119	258	59	24	31	276	584	171	55
VII) 271–390 (12)	X̄	5.72	6.09	6.69	6.89	968	421	454	524	191	86	43	11	1192	487	241	56
	σ	0.95	0.87	0.60	0.41	1526	159	189	157	360	69	30	11	1927	380	295	37
VIII) 391–430 (5)	X̄	3.81	4.56	5.75	5.39	1064	863	747	853	497	153	164	34	4990	4148	2189	2621
	σ	0.19	0.88	1.20	1.20	348	106	248	423	354	86	123	38	2011	2644	3183	2800
IX) 431–460 (6)	X̄	5.43	5.47	6.55	6.36	984	842	699	345	169	116	22	7	1098	2066	306	197
	σ	0.96	1.38	0.40	0.28	870	874	729	243	174	126	14	5	1620	2584	390	191
X) 461–600 (3)	X̄	6.01	5.94	6.73	6.70	681	534	432	410	54	57	17	5	1405	787	223	83
	σ	0.36	0.69	0.38	0.51	520	167	96	172	29	35	16	1	1009	987	343	63
Total (64)	X̄	5.12	5.20	6.65	6.30	1469	1417	1214	1084	245	104	78	17	1855	1665	666	620
	σ	0.63	0.54	0.70	0.72	950	1053	918	810	169	36	70	14	1777	1514	853	1120

[a] Number of measurements for each period.
[b] X̄ Average.
[c] σ standard deviation.

Table 2
Hydraulic and loading rate (average) parameters applied at the WETWINE plant.

Period	Q in (L/d)	VLR HUSB ($g/m^3 \cdot d$)		HRT HUSB	SLR VF ($g/m^2 \cdot d$)		SLR HF (g/m2·d)		HLR (VF, HF)[a]
		TSS	COD	h	TSS	COD	TSS	COD	($L/m^2 \cdot d$)
I	1804 ± 723	190 ± 159	3080 ± 1797	20.0 ± 2.7	7.6 ± 3.9	157.5 ± 93.2	9.2 ± 11.6	69.5 ± 75.5	60.1 ± 24.1
II	827 ± 279	192 ± 205	3436 ± 1526	43.8 ± 2.3	5.0 ± 3.4	160.3 ± 71.6	2.4 ± 2.9	17.7 ± 13.5	27.6 ± 9.3
III	2193 ± 1076	1114 ± 627	13,674 ± 3339	16.5 ± 5.0	41.8 ± 18.8	692 ± 136	12.1 ± 4.2	290.1 ± 39.0	73.1 ± 35.9
IV	296 ± 145	145 ± 149	762 ± 407	122.4 ± 0.6	8.9 ± 7.4	55.1 ± 25.1	1.5 ± 1.0	5.0 ± 4.0	9.9 ± 4.8
V	991 ± 1021	355 ± 479	2989 ± 829	36.6 ± 1.3	9.5 ± 8.1	144.1 ± 46.4	5.6 ± 5.7	6.0 ± 5.6	33.0 ± 34.0
VI	3056 ± 3505	277 ± 224	790 ± 740	11.9 ± 1.1	9.7 ± 8.4	42.1 ± 34.4	13.4 ± 13.7	28.5 ± 33.0	101.9 ± 116.8
VII	936 ± 504	55 ± 51	381 ± 306	38.7 ± 0.5	2.1 ± 1.5	13.4 ± 11.9	1.4 ± 1.8	5.6 ± 3.8	31.2 ± 16.8
VIII	1303 ± 1396	341 ± 263	4642 ± 4871	27.8 ± 7.4	7.3 ± 7.7	241 ± 298	5.6 ± 5.1	211.5 ± 420.0	43.4 ± 46.5
IX	861 ± 257	107 ± 107	721 ± 1102	42.1 ± 1.7	3.5 ± 4.3	57.6 ± 69.4	0.7 ± 0.4	8.0 ± 9.7	28.7 ± 8.6
X	3143 ± 3909	82 ± 83	1209 ± 944	11.5 ± 1.4	4.2 ± 4.0	57.4 ± 52.7	1.0 ± 0.9	12.4 ± 18.7	104.8 ± 130.3
Total	1541 ± 978	286 ± 309	3168 ± 3968	37 ± 32	10.0 ± 11.5	162.0 ± 199.2	5.3 ± 4.8	65.4 ± 101.3	51.4 ± 32.6

[a] Considering that evapotranspiration was negligible at the VF, and that the VF and HF steps had the same area, the HLR is the same for the VF and HF units (figures shown), while the HLR for the entire system corresponds to these figures divided by 2).

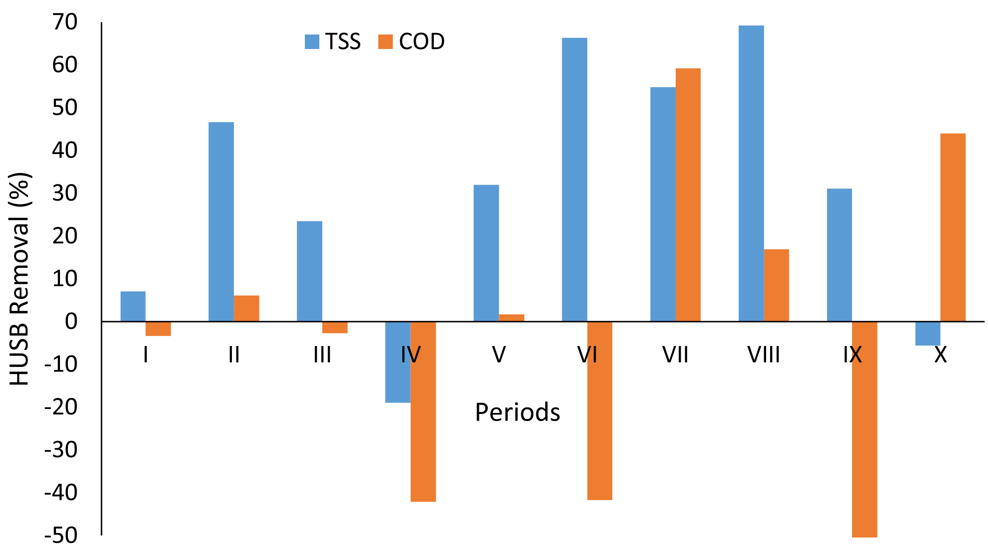


Fig. 2. HUSB Removal.

responsible for the formation of methane gas in the system is highly sensitive to pH values and it also affects the anaerobic degradation processes of organic matter (Saeed and Sun, 2012), Methanogenic bacteria can only survive at pH values above 6.5 (Almuktar et al., 2018). As a result, the anaerobic degradation process will not be completed, if the pH value is not in that range, leading to volatile fatty acid accumulation in the system and a subsequent drop in the pH value. A low pH also limits methanogen viability in constructed wetland media as reported by Cooper et al. (1996) and Vymazal (1999).

Electric conductivity (EC), did not present a considerable change of EC along the CW. and the ec values measured at the influent and effluent are similar (1468 ± 951 μS/cm and 1417 ± 1053 μS/cm respectively).

The results of this study are in accordance with the results presented by De la Varga et al. (2013a) in a previous work on the application of HUSB for the treatment of wastewater from the wine industry. In the study, De la Varga et al. (2013a), stated that the HRT in the HUSB reactor ranged between 19 and 28 h, while in the WETWINE plant, except for period IV, the HUSB operated at a HRT of 28 ± 13 h. Regarding VLR_{TSS} in both studies concentrations, ranged from 0.3 to 1.1 kg $TSS/m^3 \cdot d$ for high loading periods (Table 2). For COD for the WETWINE plant VLR_{COD} the loading to the digester averaged 3.2 ± 4.4 kg $COD/m^3 \cdot d$, higher thant the one reported by De la Varga et al. (2013a) of 2.4 ± 1.6 kg $COD/m^3 \cdot d$. The VLR_{COD} was particularly high during period III, when it reached 14Kg $COD/m^3 \cdot d$. In the study by De la Varga et al. (2013a), the HUSB removed on average 76% TSS and 26% COD, somewhat better results than those found for the WETWINE project, but that might be justified by the lower TSS and higher COD concentrations in the influent to the WETWINE plant.

According to De la Varga et al. (2013a), the HUSB reactor should minimize methanogenic activity while enhancing hydrolysis and acidification of particulate matter to reach high volatile fatty acid concentrations in the effluent implies further elimination of particulate matter. Therefore, a HUSB reactor is mainly used to treat wastewater with a high suspended solid concentration to solubilize particulate matter and to increase the removal of easily biodegradable organic matter in the wastewater. The type of substrate, influent concentration, temperature, HRT and solid retention time are the main operational parameters that define the methanogenic or non-methanogenic conditions of an anaerobic system (Álvarez et al., 2003; Álvarez et al., 2008). In spite of the long HRT applied in some of the testing periods, De la Varga (2014) reported zero methanogenic activity of accumulated solids, which was attributed to the low pH.

3.3. Performance and treatment efficiency of the VF

Fig. 3 shows the TSS and COD removal in the VF and HF units as well as in the overall system (including HUSB). The COD removal was generally high in the VF that achieved average values ranging from 47% to 96% for the different operation periods. Nevertheless, the TSS removal in VF varied, from 32 to 81% in most of the periods, but with

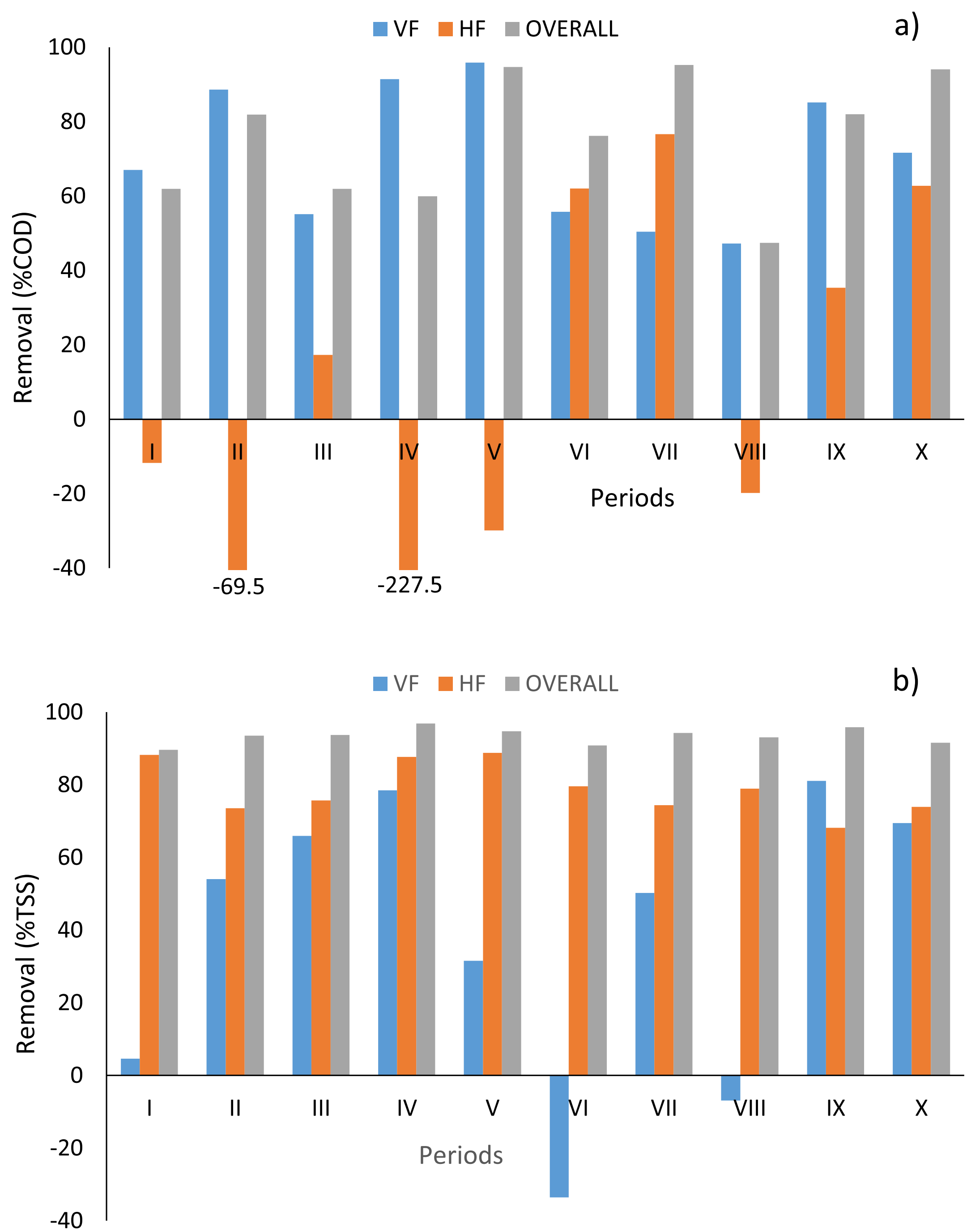


Fig. 3. COD (a) and TSS (b) removal efficiency in the VF and HF units and in the overall system.

very low removal or even negative removal (increase of COD) in periods I, VI and VIII.

The effect of the following variables: COD influent concentration (COD_{in}), SLR_{COD}, HLR, TSS influent concentration (TSS_{in}) and SLR_{TSS}, on the COD removal efficiency (% COD_r) in VF was evaluated. None of these variables correlates with % COD_r. The % COD_r increased with the SLR up to values of 100–160 g COD/m^2•d and decreased subsequently for higher SLRs (Fig. 4a). However, multiple regression analysis of all data ($n = 10$, period means) provides a good adjustment for % COD_r as a function of COD_{in} and SLR_{COD} ($R^2 = 0.70$, $R^2_{adj} = 0.62$) at a significance level of $p < 0.01$ for the two variables. According to this model, the two variables explain 70% of the variation of COD_{in} removal in the VF. The introduction of HLR or SLR_{TSS} does not help as they decreased the R^2_{adj} value and resulting in high p values.

In the case of low SLR (<100 g COD/m^2•d), the VF reached % COD_r values higher than 80% (85–96%) when the COD_{in} was greater than 4000 mg/L. When the VF operates at low SLR and influent COD concentrations less than 2000 mg/L, the percentage COD_r drops to values around 60% removal. The data analysis suggests that the % COD_r increases with COD_{in} ($R^2 = 0.74$, $n = 8$) whenever SLR is less than 200 g COD/m^2•d. This fact explains that the % COD_r was low in the periods III and VIII (55% and 47%, respectively) because of the very high SLR of 692 ± 136 g COD/m^2•d (III) and 241 ± 297 g COD/m^2•d (VIII) applied.

The HLR showed a limited effect on % COD_r ($R^2 = 0.22$) decreasing

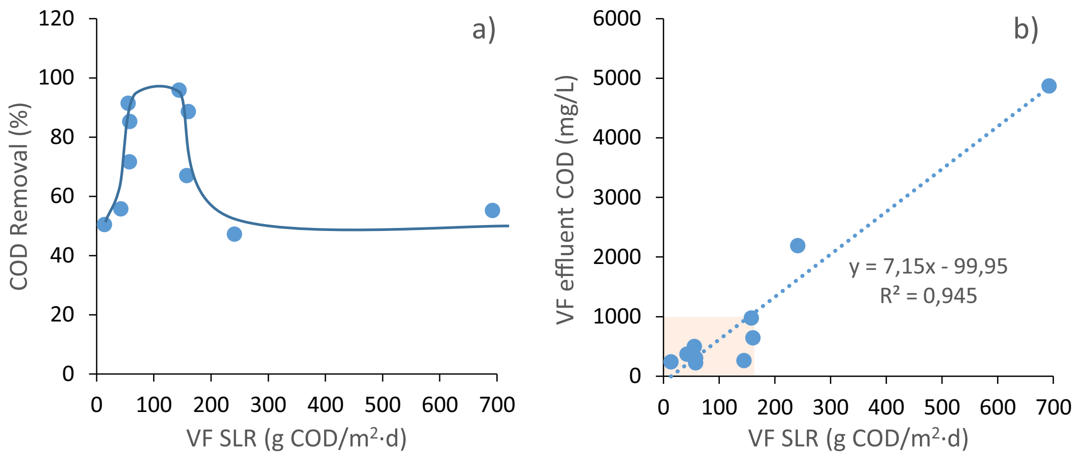


Fig. 4. Effect of SLR on COD removal (a) and effluent COD concentration (b) in VF.

slightly as the HLR increases in the range of 10 to 105 L/m^2•d. In addition, no correlation was found between % COD_r with TSS_{in} and $SLR_{TSS.}$ However, a correlation existed between COD effluent and SLR (R^2 = 0.94, p < 0.01). This correlation allows us to estimate the maximum SLR value in order to ensure a maximum effluent COD concentration (COD_{ef}). Effluent COD from the VF would be below 1000 mg/L if the SLR is below 160 g COD/m^2•d (Fig. 4b). The effect of HRT on % COD_r was studied and it was observed that it had no significant effect (R^2 = 0.18, p > 0.1). The data obtained in this work show that the efficiency of the VF is mostly affected by the SLR_{COD} and COD_{in}.

As for the % CODr and to explain the mechanisms of %TSS_r in VF a multiple regression analysis was made. Regarding the % TSS_r in the VF, none of the three variables (TSS_{in}, SLR_{TSS} and HLR), taken separately or in combination show correlation (R^2 < 0.22, p > 0.1). Neither does HRT show any effect on % TSS_r (R^2 = 0.18, p > 0.1).

Therefore, the efficiency of TSS removal in the VF is due to other mechanism such as filtration. In particular, it was found that the TSS_{ef} concentration is related to TSS_{in} by a logarithmic function (R^2 = 0.67, p < 0.01). According to the correlation (equation not shown), it was observed that the TSS_{ef} increased up to 200 mg/L with the increase of TSS_{in} concentrations up to 300–400 mg/L, while the TSS_{ef} remained almost constant (200–220 g/L) for TSS_{in} values over 400 mg/L.

Regarding the pH evolution, the VF unit was able to neutralise the typical slightly acidic effluent waters from wineries. The influent of VF had an average value of 5.2 ± 0.5 and when it left the VF presented an average value of 6.7 ± 0.7. This is explained by the removal of acidic components entering the VF such as volatile fatty acids, as suggested by the high COD removal registered. The VF also contributed to the decrease in EC, obtaining an effluent with an average EC of 1214 ± 918 µS/cm, lower than the average influent value.

3.4. Performance and treatment efficiency of the HF

The HF unit showed low removal of COD in the first part of the study (periods I until V) and medium to high COD removal in the second part (periods VI until X) (Fig. 3). Besides, the efficiency of HF on TSS removal remained always high, with average removal between 68 and 89%.

The effect of SLR_{COD} on % COD_r was studied and it was observed that the % COD_r decreased for the load was above 30 g COD/m^2•d (periods I, III and VIII) and also during the low loading periods (periods II, IV, V and IX). In the other periods such as VI, VII, and IX the performance was high (62–77% COD removal), corresponding to a SLR in the range of 6 to 28 g COD/m^2•d.

None of the parameters COD_{in} or SLR_{COD} were individually correlated with % COD_r and no correlations were obtained using multiple regression (R^2 < 0.4, p > 0.05). Only when the two high load periods are eliminated (period III and VIII, n = 8), a good adjustment for the multiple regression of % COD_r versus COD_{in} and SLR (R^2 = 0.71, p < 0.05) is obtained. Under these conditions, the model states that the % COD_r in HF increases with the increase of SLR_{COD} as well as with the decrease of the COD_{in}.

The behaviour of % COD_r in the HF unit can be explained by the absence of true stationary state and the high loads applied, which seem to be excessive for the HF. It should be considered that in periods of high load there is a retention and accumulation of organic matter in the HF bed that was not mineralized. This affected the subsequent periods of low load, generating negative % COD_r removal values. If we consider the three groups of periods indicated in Fig. 5, it allows us to obtain a correlation between % COD_r and SLR for those periods (n = 4) of low and medium load which are not affected by previous periods of high loading. An equation for the variation of the efficiency as a function of SLR under steady-state conditions was obtained (Eq. (1)). Therefore, % CODr would decrease below 50% for SLR greater than 27 g COD/m^2•d, and it would reach a zero value for an SLR of 64 g COD/m^2•d.

$$COD_r\ (\%) = 86.8–1.36\ x\ SLR\ \left(g\ COD/m^2{\cdot}d\right) \quad (1)$$

A significant effect of HRT on % CODr was found (R^2 = 0.75, p < 0.1), which is on line with the study by Akratos and Tsihrintzis (2007). They studied the relationship between HRT and COD removal efficiency snd heir results show that as the HRT decreases, the effluent COD concentration will increase. These results were confirmed by Trang et al. (2010), who observed the reduction in organic matter and nitrogen removal efficiency with the reduction of hydraulic retention time in their system due to less contact time of contaminants in the wetland resulting in low effluent quality for reuse purposes in the agricultural sector. The drop-in removal efficiency was observed in chemical oxygen demand and total suspended solids as well. The hydraulic retention time is one of the few operational factors, which can be controlled in wetland systems (Almuktar et al., 2018).

In relation to the removal of TSS, high % TSS_r values were found for the HF (Fig. 3b). The data analysis shows that the % TSS_r increases with TSS_{in} (R^2 = 0.57, p = 0.01), with values reaching 70% to 90%. An increase of the HLR values led to a slight decrease of the % TSS_r. Regarding the SLR_{TSS}, the % TSS_r initially increased with the SLR_{TSS} up to 8 g SST/m^2•d (Fig. 5b). However, when the values of SLR_{TSS} ranged between 8 and 14 g SST/m^2•d, the % TSS_r decreased although this effect was small and statistically not significant. Multiple regression indicates that none of these variables added to the influent concentration improves the correlation already indicated (causing the reduction of R^2_{adj} and p > 0.1). Besides, the HRT had no significant effect on % TSS_r (R^2 = 0.10, p > 0.1).

In summary, the obtained data (Fig. 5) show that the HF is able to

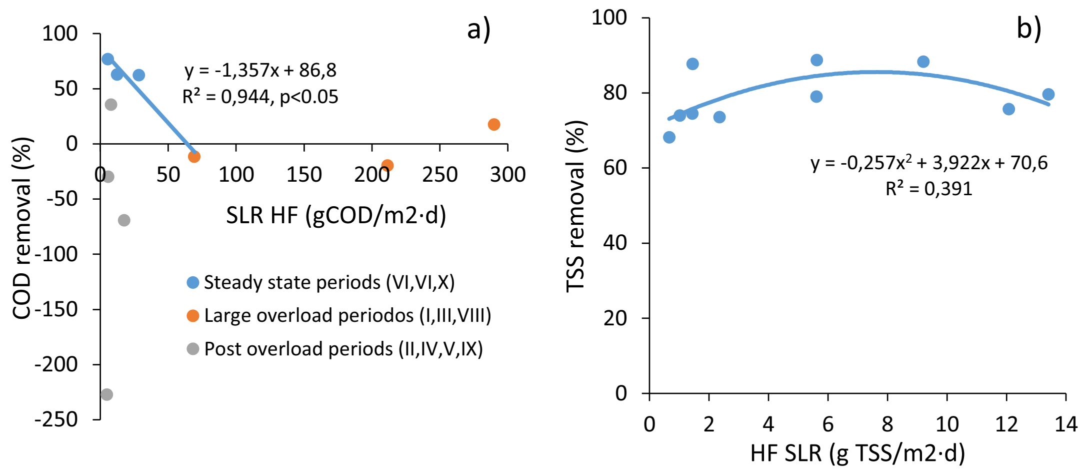


Fig. 5. Model interpreting the COD removal efficiency (a) and TSS removal (b) as a function of SLR in HF unit.

ensure an effluent with a COD removal of over 50% when the SLR applied is lower than 30 g COD/m^2·d, increasing the efficiency as the SLR become lower. In the case of TSS, the HWC obtain removal percentages higher 80% if the SLR applied is lower 8 g TSS/m^2·d. Taking into account the fact that the VF showed low solids retention, in order to ensure the discharge limit value of 35 mg TSS/L in the final effluent (reclaimed water quality requirements for agricultural irrigation, REGULATION (EU) 2020/741 and for water reuse, RD 1620/2007), the effluent concentration of VF cannot be over 175 mg/L. This means that the concentration of TSS in the influent of VF, that is, in the HUSB effluent, should not exceed 200 mg/L.

The HF produced effluent with a neutral pH values and able to reduce the conductivity value in the final effluent further. This is an improvement due to the fact that a high EC is a common problem in wine cellar water discharges. For example, in period III the effluent of HUSB presented a value of 3073 μS/cm and the final effluent (after VF and HF) was 1827 μS/cm. It was found that the greatest effect on the EC decrease is due to HF. The final effluent presents an average values of pH = 6.3 ± 0.70 and EC = 1084 ± 810 μS/cm.

If the treated effluent is to be reused for irrigation, pH and EC values are very important (SAR index). If pH is very low, the irrigated soil can become acidic, resulting in the mobilization of the nutrients and other microelements in the soil that will result on limited plant growth and long term productivity. On the other hand, for water with high pH values, the media will be basic, which will hinder crops ability to uptake the necessary elements from the soil, resulting in growth stunting and consequently very low productivity as reported by Almuktar et al., 2017). Therefore, the recommended pH value for irrigation water should be in the range of 6 to 8 (Almuktar et al., 2018).

3.5. *Effect on nitrogen and inorganic compounds*

When compared to other similar reported study (Serrano et al., 2011), the concentration of nitrogen and phosphorus compounds in the WETWINE wastewater were low, with an average nitrogen concentration at the influent of 22.9 ± 23.5 mg TN/L), mainly in form of ammonium (15.9 ± 20.4 mg NH_4^+-N/L) and the rest as organic nitrogen. The actual loading to the system was 0.4 mg NH_4^+-N/m^2·d (or 0.6 mg TN/m^2·d) 0.14 mg P/ m^2·d on average. The VF unit reached 57% ammonia removal on average, while the HF effectively removed nitrite (87%) and nitrate (97%). Total nitrogen removal reached 62% in the overall system, where at the final effluent the concentration was 7.0 ± 10.6 mg NH_4^+-N and 8.6 ± 11.9 mg TN/L. Regarding phosphorus compounds, the influent concentration was 5.5 ± 4.4 mg TP/L of which phosphates were 4.8 ± 4.0 mg PO_4^{3-}-P/L, and the overall removal reached 12% TP and 38% phosphates.

Plants have an important role in CWs, directly affecting wastewater quality by improving removal processes and consumption of phosphorus, nitrogen, and other elements (Ko et al., 2011; Ong et al., 2010). Moreover, antibiotics (Liu et al., 2013), nutrients (Scholz, 2006, 2010; Vymazal, 2007), and heavy metals (Ha et al., 2011) may accumulate in wetland plants.

However, ammonia removal was usually affected by oxygen transfer rates. Vertical flow constructed wetlands (VF) achieve a high rate of oxygen transfer (Li et al., 2015). Initially, the applied wastewater in the wetland system will be distributed homogeneously over the entire surface of the bed and then infiltrate through the system by gravity through an unsaturated bed, filled with oxygen (Stefanakis et al., 2014). As water trickles down the filter, air trapped in the pores is available to support aerobic processes as wastewater passes through the wetland media. The WETWINE VF, the wastewater is applied sequentially in pulse cycles so that there are resting period in between the loading pulses, allowing the recharge of O_2 of the bed and leading to a high rate of oxygen transfer in the system. Vertical flow constructed wetlands can remove chemical and biochemical oxygen demand as well as retain TSS from wastewater as nitrifying effectively (Brix and Arias, 2005). In the horizontal flow system, the treatment of wastewater moves horizontally through the system substrate, in contact with the plant roots, and rhizomes towards the system outlet (Vymazal, 2009). In this system, the treatment of wastewater, occurs while water travels below-ground in a water saturated bed and the combination of biological, chemical and physical process as wastewater pass through aerobic, anaerobic, and anoxic zones. (Kadlec and Wallace, 2008; Vymazal, 2014). Oxidized nitrogen compounds are effectively removed in anaerobic and anoxic environments predominating in the HF unit.

Among other inorganic compounds (F^-, I^-, Cl^-, Br^-, SO_4^{2-}, Li^+, Na^+, K^+, Ca^{2+} and Mg^{2+}), only the presence of potassium was relevant. The average concentrations of K^+ in the influent was 393 ± 411 mg/L. The average removal percentage was 48%. The high presence of K^+ in the wastewater of WETWINE plant was probably due to the type of soil in the area where the pilot plant is located. The soil of Tomiño is rich in minerals like K^+, Ca^{2+} and Mg^{2+} but in the case of the WETWINE plant only the presence of K^+ was significant. In fact, calcium and magnesium increased they concentration in the final effluent. High removal rates were observed for sulphate (87%) and Li^+ (65%) while the removal of Cl^- and Na^+ was about 25%. Ions removal contributed to a total EC decrease of 26% (Table 1).

3.6. *The efficiency of the overall WETWINE plant*

A statistical analysis of the data was also performed to assess the overall efficiency of the WETWINE plant. The average global removal of

COD was variable although generally high (48–95%). The average value of % COD_r reached 76%. No independent variables could be correlated on its own with the variation in COD_r ($R^2 < 0.15, p > 0.1$). Similarly, no combinations of variables could be adjusted to the data ($R^2 < 0.2, p > 0.3$). The global removal of TSS varied in the range of 90–97%. The average % TSS_r value was 93%. This global % TSS_r is only affected by HLR so that the % TSS_r decreases when HLR is reduced ($R^2 = 0.67, p = 0.01$). The lower values of % TSSr are obtained at low values of TSS_{in} but the correlation is not significant ($R^2 = 0.22$). In addition multiple correlations did not provide better results.

The average values for % COD_r and % TSS_r are similar that obtained in previous works (Serrano et al., 2011). These authors detailed that a full-scale plant consisted of a combination of anaerobic digestion reactor (AD) and CWs reached overall removal efficiencies of 96% and 79% an average for TSS and COD. However, the WETWINE plant operated at an overall SLR of 81 g COD/m^2·d higher than that of g 37 COD/m^2·d applied by Serrano et al. (2011). The higher capacity of the WETWINE plant should be attributed to the larger VF/HF area ratio which equals 1 compared to 0.17 for the Serrano et al. (2011) plant.

The optimal COD balance over a 100% influent establishes a 10% COD removal for HUSB, 81% for VF and 5.4% for HF, reaching a total removal 96. % (this is 10%, 80% and 60% CODr for HUSB, VF and HF, respectively, on a step basis). In the case of TSS, the overall balance indicates 67% removal for HUSB, 4.1% for VF and 23% for HF, reaching 94.% total removal (67%, 13% and 80% TSS_r for HUSB, VF and HF, respectively, on a step basis). Taking into account these results, we can estimate that during the harvest season the wastewater obtained has the following characteristics: 600 mg TSS and 8000 mg COD/L, the WETWINE plant would produce a treated effluent with the following characteristics: 35 mg TSS/L, 216 mg COD/L. In the case of the season out of harvest, the WETWINE plant would produce a treated effluent susceptible to be used for agricultural irrigation purposes but E.coli analysis must by done to confirm this use. Another option is to use the treated effluent for reuse in wetland recharge, aquifers, washing waters or irrigation of forests, green areas and other areas not accessible to the public. (Morari and Giardini, 2009).

3.7. Optimal operational conditions and VF/HF area ratio

The optimal operating conditions and performances were identified for each stage of the WETWINE plant to propose the optimal step combination for the hybrid CW system. The discharge limits of 35 mg TSS/L and 250 g COD/L were considered as reference according to RD 1620/2007. The analysis was carried out in both backward and forward direction. In the backward method, the final allowed concentration takes into account and the operating conditions were selected according to it. For the forward method, the influent concentrations were selected from either the average concentration for harvesting periods or the highest period concentration registered in the WETWINE project (Table 1). Thus, two different scenarios have been considered, depending on the raw influent concentration. Calculations followed in both methods by setting the percentage removal (% CODr or % TSSr, selecting the more restrictive option) in each unit and the conditions (SLR) to achieve this removal efficiency. This step was carried out based on the e data described in Sections 3.2 to 3.4. For the forward method, the optimal SLR was considered as a higher SLR that has not a sharply negative effect on the removal efficiency. The results obtained are shown in Table 3.

TSS and COD removal efficiencies in the HUSB were always considered to be the same (10% and 60% respectively) because they depend mainly on the operation characteristics such as the up flow velocity. In the same way, the TSS efficiency removal in VF was set at 13% in all cases, as it was not related to any sizing parameters.

Regarding the SLR and % CODr in the VF, different values have been considered for each scenario. The maximum SLR for high % CODr efficiency (160 g COD/m^2·d) was considered in scenarios B and C, while

Table 3
Optimal conditions and efficiency for each unit and the overall system.

Analysis procedure	Backward method		Forward method (medium conc.)		Forward method (high conc.)	
Parameter[a]	COD	TSS	COD	TSS	COD	TSS
Final eff. Conc. (mg/L)	**<u>250</u>**	**<u>35</u>**	353	30.8	476	38.4
%R HF	**60**	80	**50**	80	**50**	80
SLR HF (g/m2·d)	**20**	5.6	**30**	6.6	**30**	6,1
HFin. conc. (mg/L)	625	175	705	154	952	192
%R VF	**90**	12.5	**85**	12,5	**90**	12.5
SLR VF(g/m2·d)	**100**	3.2	**160**	6,0	**160**	2,3
VFin. conc. (mg/L)	6250[c]	200	4700	176	9519	220
%R HUSB[b]	10	**60**	10	**60**	10	74.0
Raw in. conc. (mg/L)	6944 [c]	500 [c]	**<u>5222</u>**	**<u>441</u>**	**<u>10,577</u>**	**<u>846</u>**
%R overall	96.4	93.0	93.2	93.0	95.5	95.5
Derived design parameters						
Overall SLR	73	5.3	103	8.7	115	9.2
HF HLR	32.0		43.0		31.5	
VF HLR	16.0		34.0		16,8	
Overall HLR	10.7		18.9		11.0	
Surface ratio VF/HF	2.0		1.3		1.9	

[a] Bold underlined figures correspond to the initial set conditions of the procedure, and bold figures to the parameters of the limiting step.
[b] The conditions for the HUSB system are HRT > 11 h and up flow velocity below 1 m/h.
[c] The analysis of the system behaviour indicate that higher concentration values would also meet the final discharge limit.

more conservative values were required by the backward method (scenario A, 100 g COD/m^2·d). The derived % CODr was calculated considering not only the SLR but the effect of the influent concentration. For VF, the resulting SLR_{TSS} was lower than potential efficiency limits.

COD removal was also the limiting step for all scenarios in HF. A conservative SLR was selected at 20 g COD/m^2·d (scenario A) while the maximum SLR of g COD/m^2·d was selected for scenarios B and C. The derived % CODr (equation in Fig. 5) were applied.

The results of Table 3 suggest that the permitted limits for the final discharge can be obtained by the WETWINE plant if SLR during harvesting periods were limited to 100 (VF) and 20 (HF) g COD/m^2·d together with a surface ratio of 2 for the VF respect the HF. On the other hand, even if the VF/HF area ratio was optimized, the application of the maximum optimal SLR would produce effluent concentrations higher than the permitted limit for the final discharge. In this case, the required CW surface will be approximately 30% less, but a post-treatment would be required. The results also indicate that the VF/HF area ratio must be higher than 1, and close to 2.

4. Conclusions

The WETWINE project uses a combination of a HUSB digester and CWs for effectively treat the wastewater produced along the year, with the consequent variations in flow and load and to produce a final effluent quality that can reclaimed for irrigation as well as to decrease the carbon footprint.

This study assessed the operation of the WETWINE plant followed for two years and evaluated the performance in regards to pollutant removals. In the applied conditions, the HUSB removed over 60% of TSS and 10% of COD of the overall influent. The VF removed over 81% and 13% of COD and TSS respectively of the influent. The HUSB was operated at HRT of 28 ± 13.0 h reaching an effluent with an average value of 104 ± 36 mg/L. The VF was operated at an average SLR of 10 g TSS/m^2·d and 162 g COD/m^2·d. The effluent produced had an average value of 78 ± 70 mg/L of TSS and 666 ± 853 mg/L of COD. The HF was operated at an average SLR of 5 g TSS/m^2·d and 65 g COD/m^2·d. This unit produces an effluent with high quality.

COD and TSS loading rates and influent concentration are the main parameters governing the removal efficiency of the system units. The VF reached % COD_r values higher than 80% (85–96%) if the COD_{in} is greater than 4000 mg/L and SLR below 100 g COD/m^2·d. The % COD_r increased with COD_{in} and sharply decreased at SLR higher than 160 g COD/m^2·d. On the other hand, the HF is able to ensure COD removal over 50% when the SLR applied is lower than 30 g COD/m^2·d, increasing the efficiency as the SLR become lower. In the case of TSS, the HWC obtain removal percentages higher 80% if the SLR applied is lower 8 g TSS/m^2·d while % TSS removal was usually low in the VF.

The VF unit reached 57% ammonia removal and the HF effectively removed nitrite (87%) and nitrate (97%). Total nitrogen removal reached 62% in the overall system. Indeed, Santiago Ruiz winery produces young white wine and the low value of pH is a typical issue for this kind of wine in the north of Spain. The results obtained show that the WETWINE plant increased the wastewater pH and finally, the effluent ranged from 5.8 to 7 pH units.

The results obtained show that the combination of HUSB and CWs is an attractive, robust and effective technology for the treatment of wastewaters mainly aimed at moderate sized wineries due to their capacity to treat the polluted water reaching the discharge limits, while requiring low maintenance and operating costs. Moreover, this system reaches the TSS value required by the new Regulation on minimum requirements for water reuse for agricultural irrigation. Regarding COD removal, this system would allow the reuse of water for irrigation as long as the influent does not exceed COD values of more than 2000 mg/L, however, in our study E.coli removal has not been studied and issues that must be considered in further studies.

From the results one can conclude that the combination of CWs can adapt to treat the heavy and variable load of wine industry while producing water of suitable for agricultural irrigation, since The it meets the requirements of RD 1620/2007 on the minimum requirements for water reuse in irrigation of forests, green areas and other areas not accessible to the public.

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.

Acknowledgements

This research was funded by the European Regional Development Fund (Interreg V-B SUDOE programme, WETWINE SOE1/P5/E0300). The authors are grateful for the support provided by the WETWINE consortium and the wineries involved in this study. Most of the results shown in this paper were presented in the 2019 WETPOL Conference (Aarhus, Denmark) under the oral presentation: “Constructed wetlands in wineries: WETWINE project” by Rocío Pena.

Funding

This research was funded by the European Regional Development Fund (Interreg V-B SUDOE programme, WETWINE SOE1/P5/E0300).

References

Akratos, C.S., Tsihrintzis, V.A., 2007. Effect of temperature, HRT, vegetation and porous media on removal efficiency of pilot-scale horizontal subsurface flow constructed wetlands. Ecol. Eng. 29, 173–191. https://doi.org/10.1016/j.ecoleng.2006.06.013.
Almuktar, S.A.A.N., Abed, S.N., Scholz, M., 2017. Recycling of domestic wastewater treated by vertical-flow wetlands for irrigation of two consecutive Capsicum annuum generations. Ecol. Eng. 107, 82–98. https://doi.org/10.1016/j.ecoleng.2017.07.002.
Almuktar, S.A.A.N., Abed, S.N., Scholz, M., 2018. Wetlands for wastewater treatment and subsequent recycling of treated effluent: a review. Environmental Science and Pollution Research 25, 23595–23623. https://doi.org/10.10007/s11356-018-2629-3.
Álvarez, J.A., Soto, M., 2011. Anaerobic treatment of domestic wastewater. In: Caruana, D.J., Olsen, A.E. (Eds.), Anaerobic Digestion: Processes, Products and Applications. Nova Science Publishers, Inc, Hauppauge, NY (USA), pp. 1–47.
Álvarez, J.A., Zapico, C.A., Gómez, M., Presas, J., Soto, M., 2003. Anaerobic hydrolysis of a municipal wastewater in a pilot-scale digester. Water Sci. Technol. 47, 223–230. https://doi.org/10.2166/wst.2003.0650.
Álvarez, J.A., Ruíz, I., Soto, M., 2008. Anaerobic digesters as a pretreatment for constructed wetlands. Ecololgical Engineering 33, 54–67.
APHA-AWWA-WPCF, 2005. Standard Methods for the Examination of Water and Wastewater, 21st ed. American Public Health Association/American Water Works Associa-tion/Water Environment Federation, Washington.
Brix, H., 2017. Sludge dewatering and mineralization in sludge treatment reed beds. Water. 9 (3), 160. https://doi.org/10.3390/w9030160.
Brix, H., Arias, C.A., 2005. The use of vertical flow constructed wetlands for on-site treatment of domestic wastewater: new Danish guidelines. Ecol. Eng. 25, 491–500. https://doi.org/10.1016/j.ecoleng.2005.07.009.
Cooper, P.F., Job, G.D., Green, M.B., 1996. Reed Beds and Constructed Wetlands for Wastewater Treatment. WRc Publications, Swindon, Witshire.
De la Varga, D., 2014. Depuración de aguas residuales en digestores anaerobios y humedales construidos: aplicación a la industria vitivinícola. Universidad de A Coruña, Chemistry Department.
De la Varga, D., Díaz, A., Ruiz, I., Soto, M., 2013a. Avoiding clogging in constructed wetlands by using anaerobic digesters as pretreatment. Ecol. Eng. 52, 262–269. https://doi.org/10.1016/j.ecoleng.2012.11.005.
De la Varga, D., Ruiz, I., Soto, M., 2013b. Winery wastewater treatment in subsurface constructed wetlands with different bed depths. Water Air Soil Poll. 224, 1485–1497. https://doi.org/10.1007/s11270-013-1485-5.
De la Varga, D., Soto, M., Arias, C.A., Oirschot, D.V., Killian, R., Pascual, A., Álvarez, J. A., 2016. Constructed Wetlands for Industrial Wastewater Treatment and Removal of Nutrients. In: Val, A., Campos, J.L., Mosquera, A. (Eds.), Technologies for the Treatment and Recovery of Nutrients from Industrial Wastewater. IGI-Global, Hershey, pp. 202–230.
Ha, N.T.H., Sakakibara, M., Sano, S., 2011. Accumulation of Indium and other heavy metals by Eleocharis acicularis: an option for phytoremediation and phytomining. Bioresour. Technol. 102, 2228–2234.
Kadlec, R.H., Wallace, S.D., 2008. Constructed Wetlands, second edition. Taylor & Francis Group, Boca Raton.
Ko, C.H., Lee, T.M., Chang, F.C., 2011. The correlations between system treatment efficiencies and aboveground emergent macrophyte nutrient removal for the Hsin-Hai Bridge phase II constructed wetland. Bioresour. Technol. 102, 5431–5437.
Li, C., Wu, S., Dong, R., 2015. Dynamics of organic matter, nitrogen and phosphorus removal and their interactions in a tidal operated constructed wetland. J. Environ. Manag. 151, 310–316. https://doi.org/10.1016/j.jenvman.2015.01.011.
Liu, L., Liu, Y.H., Liu, C.X., 2013. Potential effect and accumulation of veterinary antibiotics in Phragmites australis under hydroponic conditions. Ecol. Eng. 53, 138–143.
Masi, F., Rochereau, J., Troesch, S., Ruiz, I., Soto, M., 2015. Wineries wastewater treatment by constructed wetlands: a review. Water Sci. Technol. 71, 1113–1127. https://doi.org/10.2166/wst.2015.061.
Miller, R.L., Acton, C., Fullerton, D.A., Maltby, J., Campling, J., 2002. SPSS for Social Scientists. Palgrave, London.
Morari, F., Giardini, L., 2009. Municipal wastewater treatment with vertical flow constructed wetlands for irrigation reuse. Ecol. Eng. 35, 643–653. https://doi.org/10.1016/j.ecoleng.2008.10.014.
Mulidzi, A.R., 2007. Winery wastewater treatment by constructed wetlands and the use of treated wastewater for cash crop production. Water Sci. Technol. 56, 103–109. https://doi.org/10.2166/wst.2007.478.
Ong, S.A., Uchiyama, K., Inadama, D., 2010. Performance evaluation of laboratory scale up-flow constructed wetlands with different designs and emergent plants. Bioresour. Technol. 101, 7239–7244.
Pascual, A., De la Varga, D., Arias, C.A., Van Oirschot, D., Killian, R., Álvarez, J.A., Soto, M., 2016. Hydrolitic anaerobic reactor and aerated constructed wetland systems for municipal wastewater treatment – HIGHWET Project. Environ. Technol. https://doi.org/10.1080/09593330.2016.1188995.
Pedescoll, A., Corzo, A., Álvarez, E., García, J., Puigagut, J., 2011. The effect of primary treatment and flow regime on clogging development in horizontal subsurface flow constructed wetlands: an experimental evaluation. Water Res. 45, 3579–3589. https://doi.org/10.1016/j.watres.2011.03.049.
Ruiz, I., Álvarez, J.A., Díaz, M.A., Serrano, L., Soto, M., 2008. Municipal wastewater treatment in an anaerobic digester-constructed wetland system. Environ. Technol. 29, 1249–1256. https://doi.org/10.1080/09593330802296173.
Ruiz, I., Díaz, M.A., Crujeiras, B., García, J., Soto, M., 2010. Solids hydrolysis and accumulation in a hybrid anaerobic digester- constructed wetlands system. Ecol. Eng. 36, 1007–1016. https://doi.org/10.1016/j.ecoleng.2010.04.006.
Saeed, T., Sun, G., 2012. A review on nitrogen and organics removal mechanisms in subsurface flow constructed wetlands: dependency on environmental parameters, operating conditions and supporting media. J. Environ. Manag. 112, 429–448. https://doi.org/10.1016/j.jenvman.2012.08.011.
Scholz, M., 2006. Wetlands Systems to Control Urban Runoff. Elsevier, Amsterdam.
Scholz, M., 2010. Wetland Systems—Storm Water Management Control. Springer Verlag, Berlin.
Serrano, L., de la Varga, D., Ruiz, I., Soto, M., 2011. Winery wastewater treatment in a hybrid constructed wetland. Ecol. Eng. 37 (5), 744–753. https://doi.org/10.1016/j.ecoleng.2010.06.038.
Stefanakis, A., Akratos, C.S., Tsihrintzis, V.A., 2014. Vertical Flow Constructed Wetlands: Eco-Engineering Systems for Wastewater and Sludge Treatment. Newnes, Oxford.

Torrijos, V., Gonzalo, O.G., Trueba-Santiso, A., Ruiz, I., Soto, M., 2016. Effect of bypass and effluent recirculation on nitrogen removal in hybrid constructed wetlands for domestic and industrial wastewater treatment. Water Res. 103, 92–100. https://doi.org/10.1016/j.watres.2016.07.028.

Trang, N.T.D., Konnerup, D., Schierup, H.H., Chiem, N.H., Tuan, L.A., Brix, H., 2010. Kinetics of pollutant removal from domestic wastewater in a tropical horizontal subsurface flow constructed wetland system: effects of hydraulic loading rate. Ecol. Eng. 36, 527–553. https://doi.org/10.1016/j.ecoleng.2009.11.022.

Uggetti, E., Ferrer, I., Molist, J., García, J., 2011. Technical, economic and environmental assessment of sludge treatment wetlands. Water Res. 45, 573–582. https://doi.org/10.1016/j.watres.2010.09.019.

Uggetti, E., Ferrer, I., Nielsen, S., Arias, C., Brix, H., García, J., 2012. Characteristics of Biosolids from Sludge Treatment Wetlands for Agricultural Reuse. Ecol. Eng. 40, 210–216. https://doi.org/10.1016/j.ecoleng.2011.12.030.

Vymazal, J., 1999. Removal of BOD5 in constructed wetlands with horizontal sub-surface flow: Czech experience. Wat. Sci. Technol. 40, 113–138. https://doi.org/10.1016/S0273-1223(99)00456-4.

Vymazal, J., 2007. Removal of nutrients in various types of constructed wetlands. Science of the Total Environment 380, 48–65.

Vymazal, J., 2009. The use of constructed wetlands with horizontal subsurface flow for various types of wastewater. Ecol. Eng. 35, 1–17. https://doi.org/10.1016/j.ecoleng.2008.08.016.

Vymazal, J., 2013. The use of hybrid constructed wetlands for wastewater treatment with special attention to nitrogen removal: a review of a recent development. Water Res. 47, 4795–4811. https://doi.org/10.1016/j.watres.2013.05.029.

Vymazal, J., 2014. Constructed wetlands for treatment of industrial wastewaters: a review. Ecol. Eng. 73, 724–751. https://doi.org/10.1016/j.ecoleng.2014.09.03.

Annex V

Abstract

Constructed wetlands (CWs) technology is an established green multi-purpose option for water management and wastewater (WW) treatment, with numerous effectively proven applications around the world and multiple environmental and economic advantages. Their adaptability and low operation and maintenance (O&M) requirements make them a sustainable and cost-effective choice for various WW treatment applications.

CWs have been widely applied for over 50 years, initially for municipal wastewater and later for industrial and agricultural wastewater, livestock farm effluent, landfill leachate, and stormwater runoff. Industrial wastewater often requires pre-treatment due to its distinct composition. The introduction of oxygen in CWs, known as aerated CWs, enhances treatment efficiency, especially for nitrification and denitrification processes. These systems can be operated intermittently to improve total nitrogen removal. Aeration strategies can vary in intensity, making aerated CWs flexible and effective in removing nitrogen and organic matter.

The primary objective of this thesis is to investigate and optimise various factors affecting WW treatment in aerated CWs, encompassing urban and industrial WW, with the aim of enhancing the design parameters and future implementation of these systems. Additionally, the thesis seeks to evaluate the feasibility of employing the hydrolitc upflow sludge blanket (HUSB) reactor followed by CWs configuration for treating diverse sources of WW, including urban, food industry, and winery WW, and to assess the impact of design and operational parameters on treatment efficiency.

Research findings are intended to enhance the understanding of guidelines for CWs design, operation, and maintenance. Being carried out at outdoors pilot and full scale systems, the study spans several years and focuses on crucial factors such as unit performance, phosphorus removal, HUSB reactor for pre-treatment to prevent clogging risks in CWs, the influence of bed depth in aerated CWs, and treatment efficiency.

This research holds significant relevance in improving the design of efficient and cost-effective aerated CWs systems. It addresses the need for a better understanding of the internal processes involved in these systems and seeks to provide valuable performance data and information to guide the design and operation of aerated CWs.

This doctorate thesis is comprised of four papers. The studies presented in this thesis have been carried out in Spain and Denmark in two pilot plants and one full scale plant: HIGHWET, KT Food pilot plant and one full scale plant (WETWINE plant).

The first configuration (HIGHWET pilot plant) located in A Coruña, Spain (43° 19' 36.444" N 8° 24' 31.068" W) consisted of a HUSB reactor followed by four HF CWs working in parallel and receiving anaerobic pre-treated WW. HF1, HF2 and HF3 units are fitted with aeration, while the HF4 is not aerated in order to be used as a control (paper I). The second configuration (nearby Aarhus, Denmark) consisted of a combination of a HUSB reactor as primary treatment, followed by two parallel treatment trains (aerated line and non-aerated line) of hybrid CWs (VF and HF), several wells to allow controlled recirculation of treated waters and additional wells to host reactive media to remove P

before discharge (paper II). The WETWINE plant consisted of a HUSB reactor followed by two parallel unsaturated VF, and HF CWs (i.e. hybrid CWs) to treat the wastewater of Santiago Ruiz Winery (paper IV).

The plants were operated for years and wastewater samples were collected once or twice a week and analysed in the laboratory. The effect of different parameters on plant performance was analysed. The treatment efficiency of the different units was calculated.

This thesis discusses the use of CWs in various configurations for treating different types of WW, including urban and industrial WW from food and winery facilities.

The thesis explores the use of a HUSB reactor in combination with aerated CWs for WW treatment. These configurations were found to be effective in treating various types of WW.

The results obtained in Paper I suggest that the technology could be extended to serve larger populations, particularly in the range of 2000 to 5000 pe. In this range of application, septic tanks are not useful and HUSB digesters can clearly compete with Imhoff tanks and other wastewater pretreatments.

Paper III presents results showing that an aerated VF CWs was successful in treating a significantly higher loading rate, specifically from the food industry, while maintaining treatment efficiency.

In Paper IV, a combination of a HUSB digester and CWs was used to treat winery wastewater. This configuration was evaluated over two years and was found to effectively treat winery wastewater, which often exhibits variations in flow and load. The system produced a final effluent quality suitable for irrigation and helped reduce the carbon footprint of winery operations. The results indicate that the combination of HUSB and CWs is an attractive and robust technology for treating wastewater from moderate-sized wineries.

The combination of HUSB reactor with HF CWs engineered with aeration is indeed an innovative configuration, aimed at enhancing the capacity and effectiveness of CWs while potentially helping to prevent clogging.

The results of paper I, II and IV showed that the introduction of HUSB digester can help pre-treat the urban and industrial (food and winery) WW by breaking down and digesting organic matter, reducing the load of solids and preventing clogging issues in downstream HF CWs.

The HUSB unit removed 76-89 % in urban WW and 67% in food WW of TSS. This unit decreased its performance of TSS removal for winery WW. This was predictable due to the type of WW generated in this type of industry. The winery effluents are constituted by soluble and insoluble phases with low value of pH.

Aeration strategies, depth bed, removal efficiency are factors which affect the surface area required for CWs. The studies of papers I, II, III and IV demonstrate the positive impact of aeration in CWs on treatment efficiency for municipal and industrial (food and winery) WWs and the potential for reducing the required surface area. In this thesis, the required area may be reduced by a factor of 5 or more. In addition, from the results one can conclude that the combination of CWs can be adapt to treat the heavy and variable load of wine industry while producing water of suitable for agricultural irrigation. This has significant implications for the practical application of CWs technology.

The thesis discussed the use of intermittent aeration and its effect on pollutant removal efficiency. TN removal varied depending on the operating conditions and was favoured by effluent recirculation. It found that higher ammonia removal rates were achieved with longer aeration periods (5h/3h on/off), while shorter periods (3h/5h on/off) resulted in lower nitrification efficiency. Recirculation was identified as a good option to improve TN removal in aerated CWs.

Two materials were analysed to remove total phosphorus (TP): The Polonite and the Tobermorite. The Polonite achieved a consistent and relatively high TP removal rate of 56 ± 5% throughout the study. The TP removal rate in the Polonite was maintained at this level, even at a low TP loading rate of 0.2 g TP/m^2/d. This material appears to be effective at removing TP from the WW, and its performance remained stable during the study period.

Highlights the effectiveness and versatility of combining HUSB digesters and aerated CWs for the treatment of various types of WW, with a particular focus on applications in urban and industrial settings, including wineries. The research findings support the potential extension of CWs technology to larger populations and emphasize the attractiveness of this approach for treating moderate-sized winery WW while meeting regulatory requirements.

The thesis analyzes the use of the combination of HUSB and hybrid CW to treat high load WW such as the water from a winery and the water from a food industry. The results obtained show that this combination is a good alternative for the treatment of these types of WW. This hybrid system is capable of adapting to and withstanding the typical variations in load and flow of a winery. In addition, it also proved to be effective in the treatment of highly loaded WW from the food industry.

In summary, this thesis provides valuable insights into the performance and sustainability of aerated CWs for WW treatment, considering factors such as water depth, aeration regime and pollutant removal efficiency. It highlights the potential benefits of aeration in improving treatment performance and reducing land area requirements, while also emphasising the importance of optimising aeration practices for sustainability. This thesis aims to advance the understanding of how the combinations of HUSB reactor and aerated CWs can effectively treat urban and industrial WW. By optimising design parameters and considering the specific factors that affect treatment in these systems, this research can contribute to more efficient and sustainable WW treatment practices.

Resumen

La tecnología de humedales artificiales es una opción ecológica polivalente para la gestión del agua y el tratamiento de aguas residuales, con numerosas aplicaciones probadas en todo el mundo y múltiples ventajas medioambientales y económicas. Su adaptabilidad y sus reducidos requisitos de operación y mantenimiento los convierten en una opción sostenible y rentable para diversas aplicaciones de tratamiento de aguas residuales.

Los humedales construidos llevan más de 50 años aplicándose de forma generalizada, inicialmente a las aguas residuales municipales y más tarde a los efluentes industriales, las aguas residuales agrícolas, los efluentes de explotaciones ganaderas, los lixiviados de vertedero y las aguas pluviales de escorrentía. Las aguas residuales industriales suelen requerir pretratamiento debido a su distinta composición. La introducción de oxígeno en los CW, conocidos como CW aireados, mejora la eficacia del tratamiento, especialmente en los procesos de nitrificación y desnitrificación. Estos sistemas pueden funcionar de forma intermitente para mejorar la eliminación total de nitrógeno. Las estrategias de aireación pueden variar en intensidad, haciendo que los CWs aireados sean flexibles y efectivos en la eliminación de nitrógeno y materia orgánica.

El objetivo principal de esta tesis es investigar y optimizar diversos factores que afectan al tratamiento de aguas residuales en CWs aireadas, abarcando las aguas residuales urbanas e industriales, con el fin de mejorar los parámetros de diseño y la futura implementación de estos sistemas. Además, la tesis pretende evaluar la viabilidad de emplear la configuración híbrida de manta de lodos de flujo ascendente (HUSB) seguida de CWs para tratar diversas fuentes de WW, incluyendo WW urbanas, de la industria alimentaria y de bodegas, y evaluar el impacto de los parámetros de diseño y operación en la eficiencia del tratamiento.

Los resultados de la investigación pretenden mejorar la comprensión de las directrices para el diseño, funcionamiento y mantenimiento de los CW. El estudio abarca varios años y se centra en factores cruciales como el rendimiento de la unidad, la eliminación de fósforo, el reactor HUSB para el pretratamiento con el fin de prevenir los riesgos de obstrucción en las CW, la influencia de la profundidad del lecho en las CW aireadas y la eficiencia del tratamiento.

Esta investigación es de gran relevancia para mejorar el diseño de sistemas de CWs aireados eficientes y rentables. Aborda la necesidad de una mejor comprensión de los procesos internos implicados en estos sistemas y trata de proporcionar valiosos datos de rendimiento e información para guiar el diseño y el funcionamiento de los CW aireados.

Esta tesis doctoral consta de cuatro artículos. Los estudios presentados en esta tesis se han llevado a cabo en España y Dinamarca en dos plantas piloto y una planta a escala real: HIGHWET y KT Food plantas piloto y una planta a escala real (planta WETWINE).

La primera configuración (planta piloto HIGHWET) situada en A Coruña, España (43° 19' 36.444" N 8° 24' 31.068" O) consistió en un reactor HUSB seguido de cuatro CW HF trabajando en paralelo y recibiendo aguas residuales anaerobias pretratadas. Las unidades HF1, HF2 y HF3 están equipadas con aireación, mientras que la HF4 no está aireada para ser utilizada como control (artículo I). La segunda configuración (cerca de Aarhus, Dinamarca) consistía en una combinación de un reactor HUSB como tratamiento primario, seguido de dos trenes de tratamiento paralelos (línea aireada y línea no aireada) de CW híbridos (VF y HF), varios pozos para permitir la recirculación controlada de las aguas tratadas y pozos adicionales para alojar medios reactivos para eliminar el P antes del vertido (artículo II). La planta WETWINE consistía en un reactor HUSB seguido de dos CWs VF y HF no saturados paralelos (es decir, CWs híbridos) para tratar las aguas residuales de la Bodega Santiago Ruiz (artículo IV).

Las plantas funcionaron durante años y se recogieron muestras de aguas residuales una o dos veces por semana que se analizaron en el laboratorio. Se analizó el efecto de diferentes parámetros en el rendimiento de las plantas. Se calculó la eficacia de tratamiento de las distintas unidades.

En esta tesis se analiza el uso de CW en varias configuraciones para el tratamiento de diferentes tipos de aguas residuales, incluidas las aguas residuales urbanas e industriales procedentes de instalaciones alimentarias y bodegas.

La tesis explora el uso de un reactor HUSB en combinación con CWs aireados para el tratamiento de WW. Estas configuraciones resultaron eficaces para el tratamiento de distintos tipos de aguas residuales.

Los resultados obtenidos en el trabajo artículo I sugieren que la tecnología podría ampliarse para dar servicio a poblaciones mayores, en particular en el rango de 2000 a 5000 pe. En este rango de aplicación, las fosas sépticas no son útiles y los digestores HUSB pueden competir claramente con las fosas Imhoff y otros pretratamientos de aguas residuales.

El artículo III presenta resultados que demuestran que un CWs de VF aireado consiguió tratar una tasa de carga significativamente mayor, concretamente de la industria alimentaria, manteniendo la eficacia del tratamiento.

En el artículo IV, se utilizó una combinación de digestor HUSB y CW para tratar las aguas residuales de una bodega. Esta configuración se evaluó durante dos años y se comprobó que trataba eficazmente las aguas residuales de las bodegas, que a menudo presentan variaciones de caudal y carga. El sistema produjo un efluente final de calidad adecuada para el riego y ayudó a reducir la huella de carbono de las operaciones de la bodega. Los resultados indican que la combinación de HUSB y CW es una tecnología atractiva y robusta para el tratamiento de aguas residuales de bodegas de tamaño moderado.

La combinación del reactor HUSB con los CW de HF diseñados con aireación es, de hecho, una configuración innovadora, destinada a mejorar la capacidad y la eficacia de los CW, al tiempo que puede ayudar a prevenir los problemas de obstrucción típicos de estos sistemas.

Los resultados de los artículos I, II y IV mostraron que la introducción de un digestor HUSB puede ayudar a pretratar las aguas residuales urbanas e industriales (alimentarias y de bodegas) descomponiendo y digiriendo la materia orgánica, reduciendo la carga de sólidos y evitando problemas de obstrucción en las plantas de tratamiento de aguas residuales.

La unidad HUSB eliminó el 76-89% de los SST de las aguas residuales urbanas y el 67% de las aguas residuales alimentarias. Esta unidad redujo su rendimiento de eliminación de SST en las aguas residuales de bodegas. Esto era previsible debido al tipo de aguas residuales generadas en este tipo de industria. Los efluentes de las bodegas están constituidos por fases solubles e insolubles con un pH bajo.

Las estrategias de aireación, la profundidad del lecho y la eficiencia de la eliminación son factores que afectan a la superficie necesaria para los CW. Los estudios de los artículos I, II, III y IV demuestran el impacto positivo de la aireación en las CW sobre la eficacia del tratamiento de las aguas residuales municipales e industriales (alimentación y bodegas) y el potencial para reducir la superficie necesaria. En esta tesis, la superficie requerida puede reducirse en un factor de 5 o más. Además, a partir de los resultados se puede concluir que la combinación de CWs puede adaptarse para tratar la carga pesada y variable de la industria vitivinícola a la vez que produce agua apta para el riego agrícola. Esto tiene importantes implicaciones para la aplicación práctica de la tecnología de las CW.

En la tesis se analizó el uso de la aireación intermitente y su efecto en la eficacia de la eliminación de contaminantes. La eliminación de TN se vio quevariaba en función de las condiciones de funcionamiento y se observó favorecida por la recirculación del efluente. Se comprobó que se conseguían mayores tasas de eliminación de amoníaco con periodos de aireación más largos (5h/3h on/off), mientras que periodos más cortos (3h/5h on/off) daban lugar a una menor eficiencia de nitrificación. La recirculación se identificó como una buena opción para mejorar la eliminación de TN en los CW aireados.

Se analizaron dos materiales para eliminar el fósforo total (PT): La Polonita y la Tobermorita. La Polonita alcanzó una tasa de eliminación de TP consistente y relativamente alta de 56 ± 5% a lo largo del estudio. La tasa de eliminación de TP en la Polonita se mantuvo a este nivel, incluso a una baja tasa de carga de TP de 0,2 g TP/m2/d. Este material parece ser eficaz en la eliminación de TP de las aguas residuales, y su rendimiento se mantuvo estable durante el período de estudio.

Destaca la eficacia y versatilidad de la combinación de digestores HUSB y CW aireados para el tratamiento de diversos tipos de aguas residuales, con especial atención a las aplicaciones en entornos urbanos e industriales, incluidas las bodegas. Los resultados de la investigación apoyan la posible extensión de la tecnología de las CWs a poblaciones más grandes y enfatizan el atractivo de este enfoque para el tratamiento de las WW de bodegas de tamaño moderado cumpliendo con los requisitos reglamentarios.

La tesis analiza también el uso de la combinación de HUSB y CW híbrida para tratar aguas residuales de alta carga, como el agua de una bodega y el agua de una industria alimentaria. Los resultados obtenidos muestran que esta combinación es una buena alternativa para el tratamiento de este tipo de aguas residuales. Este sistema híbrido es capaz de adaptarse y soportar las variaciones de carga y caudal típicas de una bodega. Además, también demostró su eficacia en el tratamiento de las aguas residuales de alta carga procedentes de la industria alimentaria.

En resumen, esta tesis aporta información valiosa sobre el rendimiento y la sostenibilidad de las CW aireadas para el tratamiento de las aguas residuales, teniendo en cuenta factores como la profundidad del agua, el régimen de aireación y la eficacia de la eliminación de contaminantes. Destaca los potenciales beneficios de la aireación para mejorar el rendimiento del tratamiento y reducir las necesidades de superficie, al tiempo que subraya la importancia de optimizar las prácticas de aireación para la sostenibilidad. Esta tesis pretende avanzar en la comprensión de cómo las combinaciones de reactor HUSB y CW aireados pueden tratar eficazmente las aguas residuales urbanas e industriales. Optimizando los parámetros de diseño y considerando los factores específicos que afectan al tratamiento en estos sistemas, esta investigación puede contribuir a prácticas de tratamiento de aguas residuales más eficientes y sostenibles.

Resumo

A tecnoloxía de humidais artificiais é unha opción ecolóxica polivalente para a xestión da auga e o tratamento de augas residuais, con numerosas aplicacións probadas en todo o mundo e múltiples vantaxes ambientais e económicas. A súa adaptabilidade e os seus reducidos requisitos de operación e mantemento convértenos nunha opción sostible e rendible para diversas aplicacións de tratamento de augas residuais.

Os humidais contruidos levan máis de 50 anos aplicándose de forma xeneralizada, inicialmente ás augas residuais municipais e máis tarde aos efluentes industriais, as augas residuais agrícolas, os efluentes de explotacións gandeiras, os lixiviados de entulleira e as augas pluviais de escorrentía. As augas residuais industriais adoitan requirir pretratamiento debido á súa distinta composición. A introdución de osíxeno nos CW, coñecidos como CW aireados, mellora a eficacia do tratamento, especialmente nos procesos de nitrificación e desnitrificación. Estes sistemas poden funcionar de forma intermitente para mellorar a eliminación total de nitróxeno. As estratexias de aireación poden variar en intensidade, facendo que os CWs aireados sexan flexibles e efectivos na eliminación de nitróxeno e materia orgánica.

O obxectivo principal desta tese é investigar e optimizar diversos factores que afectan o tratamento de augas residuais en CWs aireadas, abarcando as augas residuais urbanas e industriais, co fin de mellorar os parámetros de deseño e a futura implementación destes sistemas. Ademais, a tese pretende avaliar a viabilidade de empregar a configuración híbrida de manta de lodos de fluxo ascendente (HUSB) seguida de CWs para tratar diversas fontes de WW, incluíndo WW urbanas, da industria alimentaria e de adegas, e avaliar o impacto dos parámetros de deseño e operación na eficiencia do tratamento

Os resultados da investigación pretenden mellorar a comprensión das directrices para o deseño, funcionamento e mantemento dos CW. O estudo abarca varios anos e céntrase en factores cruciais como o rendemento da unidade, a eliminación de fósforo, o reactor HUSB para o pretratamiento co fin de previr os riscos de obstrución nas CW, a influencia da profundidade do leito nas CW aireadas e a eficiencia do tratamento.

Esta investigación é de gran relevancia para mellorar o deseño de sistemas de CWs aireados eficientes e rendibles. Aborda a necesidade dunha mellor comprensión dos procesos internos implicados nestes sistemas e trata de proporcionar valiosos datos de rendemento e información para guiar o deseño e o funcionamento dos CW aireados.

Esta tesis doctoral consta de cuatro artículos. Los estudios presentados en esta tesis se han llevado a cabo en España y Dinamarca en dos plantas piloto y una planta a escala real: HIGHWET y KT Food plantas piloto y una planta a escala real (planta WETWINE).

A primeira configuración (planta piloto HIGHWET) situada na Coruña, España (43° 19' 36.444" N 8° 24' 31.068" Ou) consistiu nun reactor HUSB seguido de catro CW HF traballando en paralelo e recibindo augas residuais anaerobias pretratadas. As unidades HF1, HF2 e HF3 están equipadas con aireación, mentres que a HF4 non está aireada para ser utilizada como control (artigo I). A segunda configuración (preto de Aarhus, Dinamarca) consistía nunha combinación dun reactor HUSB como tratamento primario,

seguido de dous trens de tratamento paralelos (liña aireada e liña non aireada) de CW híbridos (VF e HF), varios pozos para permitir a recirculación controlada das augas tratadas e pozos adicionais para aloxar medios reactivos para eliminar o P antes da vertedura (artigo II). A planta WETWINE consistía nun reactor HUSB seguido de dous CWs VF e HF non saturados paralelos (é dicir, CWs híbridos) para tratar as augas residuais da Adega Santiago Ruiz (artigo IV).

As plantas funcionaron durante anos e recolléronse mostras de augas residuais una ou dúas veces por semana que se analizaron no laboratorio. Analizouse o efecto de diferentes parámetros no rendemento das plantas. Calculouse a eficacia de tratamento das distintas unidades.

Nesta tese analízase o uso de CW en varias configuracións para o tratamento de diferentes tipos de augas residuais, incluídas as augas residuais urbanas e industriais procedentes de instalacións alimentarias e adegas.A tese explora o uso dun reactor HUSB en combinación con CWs aireados para o tratamento de WW. Estas configuracións resultaron eficaces para o tratamento de distintos tipos de augas residuais.

Os resultados obtidos no traballo artigo I suxiren que a tecnoloxía podería ampliarse para dar servizo a poboacións maiores, en particular no rango de 2000 a 5000 pe. Neste rango de aplicación, as fosas sépticas non son útiles e os digestores HUSB poden competir claramente coas fosas Imhoff e outros pretratamientos de augas residuais.

O artigo III presenta resultados que demostran que un CWs de VF aireado conseguiu tratar unha taxa de carga significativamente maior, concretamente da industria alimentaria, mantendo a eficacia do tratamento.

No artigo IV, utilizouse unha combinación de digestor HUSB e CW para tratar as augas residuais dunha adega. Esta configuración avaliouse durante dous anos e comprobouse que trataba eficazmente as augas residuais das adegas, que a miúdo presentan variacións de caudal e carga. O sistema produciu un efluente final de calidade adecuada para a rega e axudou a reducir a pegada de carbono das operacións da adega. Os resultados indican que a combinación de HUSB e CW é unha tecnoloxía atractiva e robusta para o tratamento de augas residuais de adegas de tamaño moderado.

A combinación do reactor HUSB cos CW de HF deseñados con aireación é, de feito, unha configuración innovadora, destinada a mellorar a capacidade e a eficacia dos CW, á vez que pode axudar a previr os problemas de obstrución típicos destes sistemas.

Os resultados dos artigos I, II e IV mostraron que a introdución dun digestor HUSB pode axudar a pretratar as augas residuais urbanas e industriais (alimentarias e de adegas) descompoñendo e dixerindo a materia orgánica, reducindo a carga de sólidos e evitando problemas de obstrución nas plantas de tratamento de augas residuais.

A unidade HUSB eliminou o 76-89% dos SST das augas residuais urbanas e o 67% das augas residuais alimentarias. Esta unidade reduciu o seu rendemento de eliminación de SST nas augas residuais de adegas. Isto era previsible debido ao tipo de augas residuais xeradas neste tipo de industria. Os efluentes das adegas están constituídos por fases solubles e insolubles cun pH baixo.

As estratexias de aireación, a profundidade do leito e a eficiencia da eliminación son factores que afectan á superficie necesaria para os CW. Os estudos dos artigos I, II, III e IV demostran o impacto positivo da aireación nas CW sobre a eficacia do tratamento das augas residuais municipais e industriais (alimentación e adegas) e o potencial para reducir a superficie necesaria. Nesta tese, a superficie requirida pode reducirse nun factor de 5 ou máis. Ademais, a partir dos resultados pódese concluír que a combinación de CWs pode adaptarse para tratar a carga pesada e variable da industria vitivinícola á vez que produce auga apta para a rega agrícola. Isto ten importantes implicacións para a aplicación práctica da tecnoloxía das CW.

Na tese analizouse o uso da aireación intermitente e o seu efecto na eficacia da eliminación de contaminantes. A eliminación de TN viuse quevariaba en función das condicións de funcionamento e observouse favorecida pola recirculación do efluente. Comprobouse que se conseguían maiores taxas de eliminación de amoníaco con períodos de aireación máis longos (5h/3h on/off), mentres que períodos máis curtos (3h/5h on/off) daban lugar a unha menor eficiencia de nitrificación. A recirculación identificouse como unha boa opción para mellorar a eliminación de TN nos CW aireados.

Analizáronse dous materiais para eliminar o fósforo total (PT): A Polonita e a Tobermorita. A Polonita alcanzou unha taxa de eliminación de TP consistente e relativamente alta de 56 ± 5% ao longo do estudo. A taxa de eliminación de TP na Polonita mantívose a este nivel, mesmo a unha baixa taxa de carga de TP de 0,2 g TP/m2/d. Este material parece ser eficaz na eliminación de TP das augas residuais, e o seu rendemento mantívose estable durante o período de estudo.

Destaca a eficacia e versatilidade da combinación de digestores HUSB e CW aireados para o tratamento de diversos tipos de augas residuais, con especial atención ás aplicacións en contornas urbanas e industriais, incluídas as adegas. Os resultados da investigación apoian a posible extensión da tecnoloxía das CWs a poboacións máis grandes e salientan o atractivo deste enfoque para o tratamento das WW de adegas de tamaño moderado cumprindo cos requisitos regulamentarios.

A tese analiza tamén o uso da combinación de HUSB e CW híbrida para tratar augas residuais de alta carga, como a auga dunha adega e a auga dunha industria alimentaria. Os resultados obtidos mostran que esta combinación é unha boa alternativa para o tratamento deste tipo de augas residuais. Este sistema híbrido é capaz de adaptarse e soportar as variacións de carga e caudal típicas dunha adega. Ademais, tamén demostrou a súa eficacia no tratamento das augas residuais de alta carga procedentes da industria alimentaria.

En resumo, esta tese achega información valiosa sobre o rendemento e a sustentabilidade das CW aireadas para o tratamento das augas residuais, tendo en conta factores como a profundidade da auga, o réxime de aireación e a eficacia da eliminación de contaminantes. Destaca os potenciais beneficios da aireación para mellorar o rendemento do tratamento e reducir as necesidades de superficie, á vez que subliña a importancia de optimizar as prácticas de aireación para a sustentabilidade. Esta tese pretende avanzar na comprensión de como as combinacións de reactor HUSB e CW aireados poden tratar eficazmente as augas residuais urbanas e industriais. Optimizando os parámetros de deseño e considerando os factores específicos que afectan o tratamento nestes sistemas, esta investigación pode contribuír a prácticas de tratamento de augas residuais máis eficientes e sostibles.