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Attenuation mechanisms and key parameters to enhance treatment performance in vegetation filters: A review

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ABSTRACT

In times when environmental concerns are on the rise and the search of ways to reduce waste generation and to create a circular economy is booming, Nature Based Solutions (NBSs) play a very important role. Vegetation Filters (VFs) are a type of Land Application System (LAS) in which wastewater is used to irrigate a forestry plantation to treat the water and produce biomass. VFs show multiple benefits that render this technology a suitable solution for wastewater treatment, especially for scattered populations or isolated buildings that lack of connection to sewer systems. This review aims to provide a comprehensive state of the art of VF implementation, highlighting the do's and don'ts for a successful performance focusing on those factors that are essential to water treatment. Results show that VFs have a great treatment capacity when all involving factors are considered, and their efficiency tends to increase with time, as the VF develops and "gets older". Indeed, the presence of fine-textured soils, the selection of a proper vegetation species, the use of pre-treated wastewater and a water balance-based irrigation schedule alternating wetting and -drying cycles are all factors that help to achieve the best performance. However, it is necessary to design and follow a simple but rigorous operation and maintenance schedule to avoid system failure, which could lead to NO₃–N leaching towards groundwater.

1. Introduction

The land application of wastewater is an old practice that was already used in ancient Greece (Metcalf and Eddy, 2003), and there are well documented cases of this type of systems implemented during the 16th and 17th centuries in Germany and Scotland (Crites et al., 2006; Hartman, 1975). However, the use of land application systems (LASs) reached its peak during the 19th century as an alternative to direct discharge of untreated wastewater into rivers (Metcalf and Eddy, 2007). By the end of the 19th century, it was clear that LAS was a valid purification method whose performance was strictly depending on the applied waste load. Indeed, the successful operation of the system could have failed when application rates overwhelmed the treatment capacity (Jewell and Seabrook, 1979). During the 20th century, several factors led to the progressive abandonment of these systems in favour of the implementation of the current conventional treatment methods. Among these factors, those more relevant are (Jewell and Seabrook, 1979; Metcalf and Eddy, 2003):

- The increase of the population in the cities generated large volumes of wastewater that could not be treated by LASs anymore.
- The industrial activity growth led to changes in urban wastewater compositions due to the introduction of a multitude of organic and inorganic components such as metals, industrials, surfactants, etc.

However in scatter populations or where land is not a limitation, there are still several examples of LAS implementations in Europe and, especially in some areas of United States (Jewell and Seabrook, 1979). Nowadays, environmental concerns are on the rise, and the search of ways to reduce waste generation and to regenerate them as a resource is booming (European Commission, 2018). In line with the principles of sustainable development and circular economy, the recovery and

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recycling of waste and resource conservation should be promoted. Indeed, in 2018 the United Nations highlighted the potential of "Nature-Based Solutions (NBSs)" as a way to face the current challenges of water management in all sectors (WWAP (United Nations World Water Assessment Programme)/UN-Water, 2018). In decentralized areas, the technical and economic limitations of small and scattered populations hamper the effective implementation of conventional wastewater treatments. Therefore, technologies based on natural attenuation processes play an important role in these areas that have limited access to sewage networks (Ortega et al., 2011). Moreover, there are different cultures in which effluent discharges to water bodies are seen as offensive to nature. This is the case of the New Zealand's Maori culture, and one of the reasons why LASs are widely spread over the country (Selvarajah, 2005).

Vegetation filters (VFs) are a type of LAS that involves the application of pre-treated and/or treated wastewater to a vegetated soil surface, usually a forestry plantation (de Miguel et al., 2014). The combined action of soil, plants and microorganisms attenuates water contaminants as a result of physical, chemical, and biological reactions. Chemical precipitation of phosphorous with calcium present in soil and physical filtration of suspended solids during infiltration are clear examples of attenuation processes. The soil particle surface is a reactive layer able to retain contaminants by e.g. ion exchange and/or van der Waals forces. Also, living organisms play an important role in contaminant attenuation. Absorption of nutrients by vegetative species and nitrification-denitrification processes mediated by microorganisms are, among others, well-known mechanisms. The optimization and enhancement of such a natural attenuation is crucial to obtain an adequate removal efficiency under variable environmental conditions. The success of the VF technology requires a good design procedure, which includes a previous study of the environment (geology, hydrogeology, soil, climate, etc.), the selection of the appropriate plant species for phytoremediation and planting density to maximize contaminant uptake, the calculation of adequate hydraulic and contaminant loads to assure the best performance and fulfill plant water requirements. In addition, it requires a simple but rigorous operation and maintenance schedule; otherwise, the system failure would lead to groundwater pollution. Indeed, the major concern about the improper operation of a VF is the leaching of nitrate (NO₃–N), a very stable species that is rapidly produced by soil nitrification processes of ammonium (NH₄-N).

There is an increasing promotion of NBSs in EU policies to achieve more sustainable and resilient societies (European Commission, 2016). In this scenario, VFs are a great option for both research and real implementation purposes, as they are a type of NBS that brings a lot of different benefits and advantages, including some that other NBSs do not, such as biomass production and landscape and biodiversity benefits. It is of pivotal importance to gather and analyze information about previous experiences with the overarching goal of accomplishing with the best treatment performance of VFs. Thus, this review aims to provide a comprehensive state of the art of VF implementation, highlighting the do's and don'ts for a successful performance with special attention to: i) key parameters affecting the treatment effectiveness in terms of nutrients (nitrogen, N, and phosphorus, P) and organic matter (OM); ii) groundwater impacts due to NO3-N leachate; iii) improvements to achieve the best performance of the VF, including soil amendment applications; and iv) the possible application of these systems for industrial wastewater and sewage sludge application.

2. Methodology

In this review, we refer to VFs as LASs with a plantation of any type of vegetation established on a natural soil, that may contain amendments and that receives any type of wastewater or reclaimed water through a controlled irrigation practice.

To accomplish the aims exposed above, 139 among scientific and technical papers from 1966 to 2020 have been reviewed; the majority of

them have been published in the last 20 years (Fig. 1). Most papers were retrieved from the Web of Science database. Search strings used were vegetation filter OR land application system OR wastewater AND (short rotation coppice OR poplar OR willow), among others. A minor number of studies was "grey literature", i.e. from not peer-reviewed literature and were found via Google Scholar searches, using the same or similar key word combinations. Searches were performed until December 2020.

A database was created extracting the following information of each reviewed paper: type of water, type of soil, plant type and species, country in which the study was carried out, hydraulic loads, nutrient concentrations in the influent and effluent and removal percentages during the VF treatment. Only those studies that reported hydraulic loads, data concentration in the influent and effluent were used to calculate removal percentage. For those where data about removal percentages were already provided, these values were considered.

Most of the reviewed literature reporting experimental results are field scale studies, while those carried out at laboratory scale only account for 10% of reviewed papers. The studies are distributed worldwide, with research experiences from all five continents. However, Sweden (10), Spain (9), China (9) and Canada (8) are the countries where VFs have been more often investigated (Fig. 2).

Net mapping analysis was performed using the software VOSViewer (version 1.6.17, CWTS, Leiden University) and two net maps were created, both representing keywords occurrence and relationships. In the first map, a minimum keyword occurrence of 5 times was used, while this requirement was lowered to an occurrence of 3 times in the second map. As can be seen in the first map (Fig. 3a), most used keywords in the reviewed literature are strongly related and they create a tangled net, mirroring the complexity that exists in a VF. Also, when minimum occurrence is lowered to 3 (Fig. 3b), this tendency persists and, at the same time, it is also noticeable that 2 different branches appear outside of the main knot; one linked to nitrogen and its attenuation mechanisms (nitrification and denitrification); and another one linked to crop parameters (evapotranspiration and crop coefficient). These two branches reflect the importance of nitrogen transformations in VFs (hereby, we pay special attention to this topic in the review), and that there are several works focusing on the plant, respectively.

3. VF treatment efficiency and attenuation mechanisms

From a general point of view, the VFs described in the reviewed literature have high treatment efficiency. In global terms, average removal rates \pm standard deviations (from minimum to maximum values) are: 88 \pm 12% (from 48 to 99%) for Biological Oxygen Demand (BOD), 85 \pm 10% (from 46 to 99%) for Chemical Oxygen Demand (COD), 78 \pm 18% (from 30 to 100%) for Total Nitrogen (TN) and 80 \pm 27% (from –33 to 100%) for Total Phosphorus (TP). Tables 1–3 present some relevant data about the attenuation of these parameters.

The processes that occur in a VF and can affect the fate of the two main nutrients present in wastewater (N and P) are described below.

3.1. Nitrogen removal

 NH_4^+ is the main inorganic N form present in raw wastewater (Levy et al., 2011). Most of organic N (urea) contained in wastewater change rapidly to NH_3 and NH_4^+ via hydrolysis and mineralization (ammonification). In this stage if the soil and wastewater pH are relatively high (e.g. calcareous soils), some N can be removed via volatilization of ammonia during wastewater application (Duan et al., 2015; Paranychianakis et al., 2006; Tzanakakis et al., 2007). Also, the protonated form of ammonia (NH_4^+) can be adsorbed by negatively-charged clay minerals and organic fraction in soil (Phillips, 2002). Under oxic conditions, NH_4^+ is oxidized to NO_3^- via nitrification by soil nitrifying bacteria (Levy et al., 2011; Meding et al., 2001). At the same time, different amounts of NO_3^- (and also NH_4^+) can be attenuated by plant uptake processes (Levy et al., 2011). In the absence of oxygen, NO_3^- is

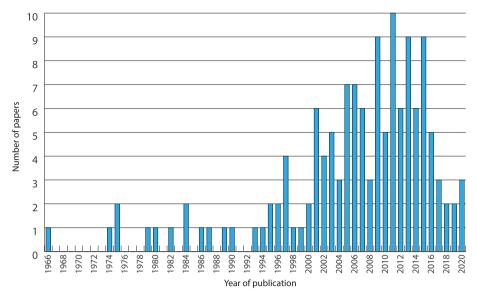


Fig. 1. Number of scientific and technical papers reviewed per year of publication.

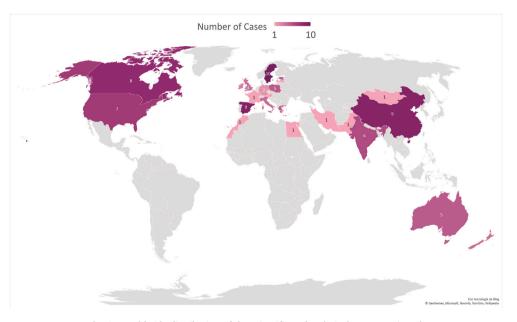


Fig. 2. Worldwide distribution of the scientific and technical papers reviewed.

used as an electron acceptor by facultative anaerobic heterotrophic bacteria (Levy et al., 2011) via denitrification thus transforming NO_3^- to N_2 . If this transformation does not occur or it occurs only partially, NO_3^- leaching can occur. The efficiency of the reactions does not only depend on the local redox conditions but also on the microbial activity and its interaction in the rhizosphere of plants. Indeed, most of the microbiota lives in the rhizosphere (Perttu and Kowalik, 1997) where root development promotes microbial activity which provides NO_3^- to the plants as an essential component for their growth.

As can be recognized, N removal pathways in a VF (Fig. 4) are quite similar to those occurred in conventional treatment plants where N is essentially removed using the nitrification-denitrification cycle by a two stage process (NH_4^+ oxidation to NO_3^- and NO_3^- reduction to N_2 gas) (Levy et al., 2011). However, the control and modulation of these processes in natural systems is challenging and, very often, the promotion of these processes fails as consequence of several factors that will be addressed later.

In a VF, climatic conditions determine the relative importance of N

transformation processes. Thus, the N uptake by plants strongly depends on their growing stages (Pandey et al., 2011) and on the season (Duan et al., 2010). Indeed, during the winter and spring seasons, denitrification plays a very important role in nutrient removal compared to plant uptake.

The average TN removal efficiency calculated from values provided by reviewed papers is 78%. The greatest removal (99.8%) was observed in a system that used pig slurry as a fertilizer in willow plantation (Cavanagh et al., 2011). The authors reported two main reasons as responsible for this high performance. First, they pointed out that during and after the application of the pig slurry, there were high rates of ammonia volatilization. Second, they claimed that during the second year, the development of willows (mostly of their root system) enhanced N uptake and decreased N amounts in the soil. Other works achieved very good performances (>90%) in terms of TN removal (Barton et al., 2005; Curneen and Gill, 2016; Dimitriou and Aronsson, 2011; Duan and Fedler, 2010; Zhang et al., 2005). The high attenuation is explained as the result of: i) high nutrient plant uptake, low permeability of subsoils,

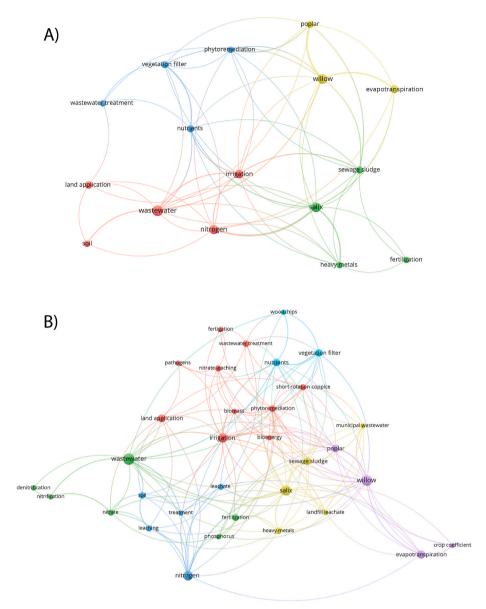


Fig. 3. Keyword's occurrence net maps for the reviewed literature. A: 5 occurrences. B: 3 occurrences.

low tendency for preferential flows in the soil and runoff dilution during the wet season (Barton et al., 2005; Curneen and Gill, 2016); ii) the pretreatment in aeration ponds, where high NH₃ volatilization occurred (enhanced by high pH and temperature values) (Duan and Fedler, 2010); iii) the proper development of willow and poplar root systems previous to irrigation and the even distribution and irrigation of wastewater (Dimitriou and Aronsson, 2011); and iv) the intermittent operation, which enhanced the oxidative-reductive environment cycles in soil, encouraging nitrification and denitrification (Zhang et al., 2005).

TN removal below 50% are observed in studies performed by Jarecki et al. (2012); Kadam et al. (2009); Kuusemets et al. (2001); Perttu and Kowalik (1997); and Rastas Amofah et al. (2012). Rastas Amofah et al. (2012) used willow plantation on coarse gravel beds to treat domestic wastewater and got NH_4^+ -N reductions of 20–30%. The authors of this study claimed that NH_4^+ attenuation occurred as the result of two main process: plant uptake and volatilization, while nitrification did not occur. Perttu and Kowalik (1997) suggested that the hydraulic loads of 4000 and 2000 mm (200 mm per week and 100 mm per week, respectively) were far too high to achieve a satisfactory performance and their final recommendation was to lower hydraulic loads to 1000 mm. In Kadam et al. (2009), the limited efficiency in terms of TN removal was

related to a low nitrification rate as a result of the presence in the effluent of organic carbon sources more susceptible to oxidation than NH_4^+ -N. In the study of Kuusemets et al. (2001), nitrification and final removals were hampered by the short retention times in the selected filter media (coarse sand and fine gravel) and the poor aeration determined by constantly keeping a water level of 20 cm above the surface.

The leaching of NO₃–N through the unsaturated zone towards the underlying aquifer and the associated surface water bodies is one of the most frequent concerns when using VFs. The excess of N, along with P, in water resources gives rise to problems such as eutrophication of rivers, lakes and coastal waters and groundwater contamination. (Cavanagh et al., 2011). In each countries and regions, the maximum level of NO₃–N that is possible to infiltrate through the vadose zone is regulated by different normatives in the framework of water reuse. The Environmental Protection Agency (EPA) establishes the maximum contaminant level for NO₃–N in drinking water to 10 mg/L (US EPA, 2006). Table 1 shows the studies that reported average NO₃–N leaching concentrations higher than the threshold established by the EPA. All of them show a great nitrification potential, but a poor denitrification efficiency.

High NO_3 -N concentrations in drainage water (i.e. effluent) is mainly controlled by: i) soil drainage capacity, which depend on its

Table 1

Relevant data about the operation of VFs with average concentration of NO₃-N in drainage water (effluent) higher than 10 mg/L.

Reference	Type of water ^a	Type of soil/ substrate	Plant species	Hydraulic load		Concentration of NO3–N in effluent		Concentration of N species in influent			
				Hydraulic loading rate	Units	NO ₃ –N mean (mg/L)	NO ₃ –N max. (mg/L)	TN (mg/ L)	TKN ^b (mg/L)	NH4–N (mg/L)	NO ₃ –N (mg/L)
Schipper et al. (1989)	Secondary WW	Clay soil + ash	Pinus radiata	2860	mm/ y	37	≈50	40.0	No data	No data	No data
(de Bustamante, 1990) ^c	Raw WW	No data	Populus euroamericana	No data	No data	≈ 20	No data	No data	No data	No data	No data
Martinez (1997)	Pig slurry	Silt soil	Lolium perenne	99.0	mm∕ y	123	339	No data	5320.0	3520.0	No data
Mantovi et al. (2006)	Pig slurry	Silty clay soil	Maize - Wheat - Sorghum	14.5	mm/ y	≈60	80.0	No data	No data	No data	No data
Watzinger et al. (2006)	Landfill leachate	Gravel and Biowaste Compost	Salix viminalis - Populus nigra	1763	mm/ y	105 ± 13	≈125	No data	No data	143 ± 25	1 ± 0.8
Dimitriou and Aronsson (2011)	Raw WW	Clay soil	Populus trichocarpa	910	mm/ y	≈10	≈20	34.7	No data	27.4	0.2
Tzanakakis et al. (2011)	Primary WW	Clay loam soil	Acacia cyanophylla	1188	mm∕ y	≈115	≈ 310	106.1	105.0	$\begin{array}{c} \textbf{89.0} \pm \\ \textbf{11} \end{array}$	1.1 ± 0.3
			Eucalyptus camaldulensis	1142	mm/ y	≈ 120	≈225				
			Populus nigra	887	mm/ y	≈95	≈185				
			Arundo donax	1091	mm/ y	≈65	≈150				
Cavanagh et al. (2011)	Pig slurry	Clayey loam soil	Salix miyabeana SX67	No data	No data	11.6	19.0	4920 mg/kg	No data	2750 mg/kg	No data
de Miguel et al. (2014)	Primary WW	Loam soil	Populus alba	808	mm/ y	40.1 ± 3.4	$\begin{array}{c} 48.5 \pm \\ 3.15 \end{array}$	154.90 ± 7.8	No data	145.8 ± 7.2	$\textbf{0.4}\pm\textbf{0.3}$
Duan et al. (2015) ^d	Dairy farm WW	Silty clay loam	Vines+ryegrass	162	mm/ y ^d	$\begin{array}{c} 20.0 \ \pm \\ 14.3 \end{array}$	No data	$\begin{array}{c} 429.7 \pm \\ 43.2 \end{array}$	No data	375.5 ± 19.4	$\textbf{6.4} \pm \textbf{2.4}$
Khurelbaatar et al. (2017)	Primary WW	Sand soil	Willow - Poplar	1780	mm/ y	≈11.1	No data	$\begin{array}{c} 56.0 \pm \\ 19 \end{array}$	No data	$\begin{array}{c} \textbf{45.0} \pm \\ \textbf{16.0} \end{array}$	No data

^a WW: wastewater.

^b TKN: Total Kjeldahl Nitrogen.

^c Data from the LAS located in Villarrubia de los Ojos (Castile-La Mancha, Spain).

^d WW was only applied from late September to late November.

texture and aggregation, as well as the presence of preferential flow paths (Duan et al., 2015; Mantovi et al., 2006; Watzinger et al., 2006), ii) soil microbiology and iii) prevailing redox conditions. Other possible causes might be the neutral or low pH values, as they reduce volatilization of NH₄–N (Duan et al., 2015; Paranychianakis et al., 2006), high concentrations of N in the applied wastewater (i.e. influent) (Cavanagh et al., 2011; Martinez, 1997; Tzanakakis et al., 2011), high hydraulic loads (Dimitriou and Aronsson, 2011), extreme meteorological conditions, irrigation during winter and washing of soil and sediments due to fluctuations of the groundwater table (Mantovi et al., 2006).

Table 2 shows the studies that reported average NO₃–N leaching concentrations lower than 10 mg/L at the last point of treatment (deepest recovered sample). The main processes behind this good performance are i) plant uptake, denitrification and/or volatilization (Curneen and Gill, 2016; Godley et al., 2005), ii) low influent N content (Christen et al., 2010), iii) organic N rather than inorganic in the influent (Barton et al., 2005), iv) presence of calcareous soils (De Bustamante, 1990), v) effect of dilution (Godley et al., 2005) and vi) the absence of fertilizer practices prior to wastewater irrigation (Mohamed et al., 2013).

3.2. Phosphorus removal

P in wastewater is mainly in the form of orthophosphates such as PO_4^{3-} , HPO_4^{2-} , $H_2PO_4^{-}$ and H_3PO_4 (Metcalf and Eddy, 2003). Through inorganic reactions, these orthophosphate species can either be sorbed by iron (Fe) and aluminum (Al) (hydro)oxides in acid soils or precipitate as calcium phosphate (Ca₃(PO₄)₂) in calcareous soils (Bouwer, 1974;

Duchaufour, 1984; Meffe et al., 2016). In conventional WWTPs, chemical methods that aim to resemble processes occurring in the natural environment use Fe, Al or calcium (Ca) salts to promote P precipitation (Levy et al., 2011).

In addition, orthophosphates can also be taken up by the plants of the VF (Barton et al., 2005; Duan et al., 2015) or biologically removed by polyphosphate-accumulating organisms (PAOs). PAOs are a type of microorganisms that, under anaerobic conditions, can take up volatile fatty acids (VFA) into poly- β -hydroxyalkanoates (PHA), using the energy generated via hydrolyzing their intracellular polyphosphate (poly-P) into soluble phosphates. Then, under aerobic conditions, they can take up orthophosphates to recover their poly-P levels, while oxidizing PHA for energy (Levy et al., 2011; Saia, 2017). Therefore, if the whole reaction pathway occurs, these bacteria can remove orthophosphates from the wastewater.

The average removal of TP calculated from values provided by reviewed papers is 80%. There is a couple of studies that reached a 100% of TP removal (Cavanagh et al., 2011; Fillion et al., 2009) and some others had efficiencies above 95% (Barton et al., 2005; Christen et al., 2010; Curneen and Gill, 2016; Li et al., 2012; Zhang et al., 2005). TP removal rates under 50% are observed in Guidi Nissim et al. (2014), Kuusemets et al. (2001), Mohamed et al. (2013), Pandey et al. (2011), and Rastas Amofah et al. (2012). In general, those studies that achieved better performances in terms of TP removal are the same that did so for N removal, and similarly those researchers that failed to obtain satisfactory TP attenuation were attaining low removals of N. In the case of Guidi Nissim et al. (2014), the extremely low TP concentrations in the applied wastewater (0.05–0.12 mg/L) hampers a clear assessment about

Table 2

Relevant data about the operation of VFs with average concentration of NO3-N in drainage water (effluent) lower than 10 mg/L

Reference	Type of water ^a	Type of soil/substrate	Plant species	Hydraulic load		Concentration of NO ₃ –N in effluent		Concentration of N species in influent			
				Hydraulic loading rate	Units	NO ₃ –N mean (mg/L)	NO ₃ –N max. (mg/L)	TN (mg/ L)	TKN ^b (mg/L)	NH ₄ –N (mg/L)	NO3–N (mg/L)
Handley and Ekern	Secondary WW	80% Kaolinitc clay	Brachiaria mutica	6.9	mm/d	0.4	1.1 ± 0.8	33.8	26.5	No data (7.4	No data (7.4
(1984)				9.6	mm/d	0.5	$\textbf{0.9} \pm \textbf{1.0}$			incluiding NO ₃ –N)	incluiding NO ₂ N)
				21.3	mm/d	1.7	5.5 ± 3.0				
				14.1	mm/d	0.7	$\textbf{2.2} \pm \textbf{1.2}$				
				29.4	mm/d	2.6	5.0 ± 4.7				
(de Bustamante, 1990) ^c	WW + Vinases (Raw WW)	No data	Populus euroamericana	No data	No data	7	No data	No data	No data	No data	No data
Kuusemets et al. (2001)	Primary WW	Constructed beds (humus over sand and gravel)	Salix viminalis - Salix dasyclados	No data	No data	≈0.4	≈2.3	≈57	No data	≈41.6	<0.25
Barton et al. (2005)	Secondary WW	Allophanic	Ryegrass	2300	mm/y	0.01	No data	15 ± 0.9	No data	5.3 ± 1.1	3 ± 0.4
	5	Gley	, ,	2100	mm/y	0.14	No data				
		Pumice		2300	mm/y	0.03	No data				
		Recent		2300	mm/y	0.06	No data				
Godley et al. (2005)	Landfill leachate	No data	Willow	70.9	mm/y	4.6	42.2	No data	No data	86.3	No data
Kadam et al. (2009)	Raw WW	No data	Green cover	42.0	mm/h	5.1 ± 6.2	No data	$\begin{array}{c} 10.8 \pm \\ 6.4 \end{array}$	5.9 ± 3.1	$\textbf{4.3}\pm\textbf{3.2}$	4.3 ± 4.7
Duan and Fedler (2010)	Secondary WW	Sandy clay - Clay loam soils	Bermuda grass	No data	No data	1.1	3.3	11.7	No data	2.38	4.56
Rastas Amofah	Primary WW	No data	Salix iminalis - Salix dasyclados	23.0	mm/d	<0.03	No data	23.8	No data	18.2	<0.7
Mohamed et al.	Pretreated WW	No data	Native vegetation	No data	No data	0.4	No data	5.3–30	1.33–20	0.24–5.2	0.01–0.3
(2010)	Pretreated WW	No data	Native vegetation	No data	No data	2.6	No data	10-38	1.12–35	0.022–6.7	0.01–0.4
	Primary WW	No data	Fruit trees and ornamental garden beds	No data	No data	2.1	No data	24–32	19–25	0.4–15	0.04–2.1
	Primary WW	No data	Lawn and fruit trees	No data	No data	2.3	No data	12–31	12-22	19–25	0.05–0.38
Guidi Nissim et al. (2014)	Polluted GW	Silt loam - Sandy clay loam	Salix miyabeana SX67	261	mm/y	4.1	12.66	No data	53.4	No data	0.19
Curneen and Gill	Primary WW	Clay soil	Salix viminalis	2.1	mm/d	1.1	No data	62.43	61.90	51.20	0.60
et al. (2012) Mohamed et al. Pret (2013) Pret Prin Guidi Nissim et al. Poll (2014) Curneen and Gill Prin	Secondary WW	-		0.9	mm/d	0.2	No data	105.14	104.50	54.70	0.70
	-			0.4	mm/d	1.0	No data	84.11	40.60	12.10	48.60
				0.4	mm/d	1.1	No data	84.11	40.50	12.00	48.60
				0.8	mm/d	0.7	No data	93.02	46.20	18.40	43.30

^a WW: wastewater.

^b Total Kjeldahl Nitrogen.

^c Data from the LAS located in Daimiel (Castile-La Mancha, Spain) design to treat WW from wine production whose income was seasonal (high in winter).

Table 3

Relevant data about the operation of VFs with results regarding P, COD and BOD attenuation.

Reference	Type of water	Type of soil/substrate	Plant species	Hydraulic load	Removal percentages (%)				
				Hydraulic loading rate	Units	Р	P species analyzed	COD	BOD
Ouazani et al. (1996)	Pretreated effluent	Clayey sand (12-88%)	Grass	3 imes 110	mm∕ week	10.0	TP	95.0	No data
						60.0	TP	85.0	No data
						80.0	TP	95.0	No data
Ou et al. (1997)	Municipal wastewater	Natural	Populus	96.4	cm∕ year	84.2	TP	65.4	95.1
			Fruit trees, larch and Pinus	150	cm/ year	92.8	TP	81.4	86.1
Kuusemets et al. (2001)	Municipal wastewater	Natural	Willow	27.8	mm/ day	15.0 (8.0)	TP (PO ₄)	No data	75.0
(Watzinger et al., 2006)Mant et al.	Farm wastewater	Pea shingle (Artificial)	None	No data	No data	85.8	PO ₄ –P	No data	90.8
(2003)			Willow	No data	No data	90.6	PO ₄ –P	No data	90.4
	Farm wastewater + 10 ppm Cu			No data	No data	89.6	PO ₄ –P	No data	81.0
	Farm wastewater + 100 ppm Cu			No data	No data	58.4	PO ₄ –P	No data	68.0
Zhang et al. (2005)	Raw rural wastewater after sedimentation	Red clay + cinder (Artificial)	Rye grass	2 (continuous)	cm/ day	98.0	TP	82.7	No data
				2 (intermittent)	cm∕ day	98.3	TP	90.0	No data
				4 (intermittent)	cm∕ day	98.5	TP	87.0	No data
				6 (intermittent)	cm/ day	98.7	TP	92.4	No data
				8 (intermittent)	cm/ day	95.6	TP	85.0	No data
				10 (intermittent)	cm∕ day	96.7	TP	71.8	No data
Pandey et al. (2011)	Domestic wastewater	Natural	Eucalyptus + Melia + Poplar + Willow	200	mm/ week	34.0	TP	46.0	48.0
Rastas Amofah et al. (2012)	Municipal wastewater	Layers of gravel (Artificial)	Willow	23	mm/ day	23.0	TP	80.0	87.0
Li et al. (2012)	Domestic wastewater	Activated sludge + brown soil + coal slag (artificial)	Common grass	81 (6.71 g BOD/ m ² d)	mm/d	96.8	TP	94.6	No data
				81 (9.30 g BOD/ m ² d)	mm/d	94.6	TP	91.3	No data
				81 (16.80 g BOD/m ² d)	mm/d	91.8	TP	87.5	No data
				81 (20 g BOD/ m ² d)	mm/d	82.5	TP	80.3	No data
				40	mm/d	93.0	TP	85.0	No data
				65	mm/d	90.0	TP	82.0	No data
				81	mm/d	88.0	TP	81.0	No data
				125	mm/d	80.0	TP	71.0	No data
Duan et al. (2015)	Farm wastewater	Silty clay loam	Ruegrass	6	mm∕ day	94.6	TP	76.8	No data
Li et al. (2015)	Domestic wastewater	5% activated sludge + 65% meadow brown soil + 30%	Poa annua + Ryegrass	12 (continuous mode)	cm⁄ day	58.5	TP	81.4	83.8
		coal slag	· -	10 (intermittent)	cm⁄ day	77.1	TP	91.5	96.0
				8 (intermittent)	cm⁄ day	71.4	TP	92.3	96.2
				6 (intermittent)	cm/ day	71.4	TP	93.1	96.5
				4 (intermittent)	cm⁄ day	78.0	TP	92.8	96.5

its performance. However, TP contents in the soil measured before and after the study lead to the conclusion that P leached from the soil due to the low influent concentrations. Therefore, despite the fact that N leaching is the main and most frequent concern in the operation of VFs, P leaching can also be an issue (Dimitriou and Aronsson, 2011).

percentages (when available) in relation of VF operational parameters. COD and BOD data are presented as support information together with P data, as N and P are the main issues regarding contaminant attenuation in a VF while attenuation of COD and BOD is usually much easier to achieve. Works presented in Table 3 were selected because they reported for the 3 species the lowest removal percentages. However, removal

Table 3 shows interesting results regarding P, COD and BOD removal

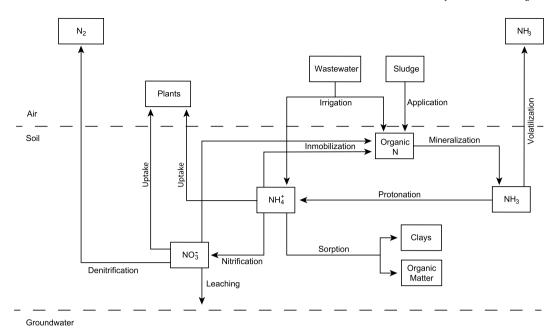


Fig. 4. Nitrogen pathways in a VF.

percentages for COD and BOD are still pretty high. Authors claims that the removals were not at their maximum due to the high hydraulic and contaminant loads, and the absence of wetting-drying cycles (Li et al., 2012, Li et al., 2012, 2015; Zhang et al., 2005). Mant et al. (2003) claimed the presence of metals such as Cu in the wastewater seems to hamper the attenuation of BOD and P. Such effects were observed when Cu-rich solution contained a 10 ppm of Cu in the case of BOD and 100 ppm of Cu in the case of P suggesting that the presence of this metal inhibited microbial activity.

4. Key parameters

4.1. Vegetation

The vegetation types and genus percentage distributions of VFs is presented in Fig. 4. Among vegetation, trees are the most used in VFs, followed by herbaceous plants and subordinately by crops. Whereas, less abundant are the studies that use a combination between these vegetation types (Fig. 5a). Among trees, willows and poplars are the species most frequently employed (Fig. 5b). The reason is mainly related to their use for biomass production coppices and to certain characteristics that make them suitable for such systems. The wider spread of willow compared to poplars can be explained by i) the number of studies carried out in Sweden and Canada, where climate conditions are more favorable for willows than for poplars, and ii) the fact that studies investigating poplars in VFs usually also test willows for comparison.

The presence of plants in a VF provides several benefits: i) it contributes to the attenuation of contaminants, such as N, P, OM or even trace metals (Aryal and Reinhold, 2015; Dimitriou and Aronsson, 2011; Guidi Nissim et al., 2014; Hasselgren, 1998; Ouazzani et al., 1996; Tsiknia et al., 2013) and ii) it may generate an economic value through sustainable forestry and timber production (Cozzi et al., 2015; Dimitriou and Aronsson, 2005; Farahat and Linderholm, 2015; Hasselgren, 1998; Tzanakakis et al, 2009, 2012).

For instance, Mant et al. (2003) described how the presence of *Salix viminalis* in a gravel hydroponic system increased the treatment efficiency by 12% of N, 5% of P and 18% of K. They suggested that this increase might be due to nutrient uptake while the willow is growing and/or the increased reactive surface area due to biofilm growth stimulated by the roots. Khurelbaatar et al. (2017) showed another example of improved performance of these systems when willows or poplars are planted. In their study, the planted treatment systems had smaller

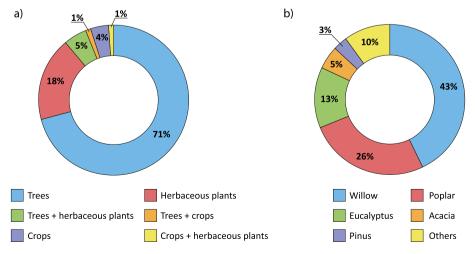


Fig. 5. A) Vegetation types used in the VFs described by the reviewed papers. B) Tree genera used in these VFs.

amounts of NH₄–N and NO₃–N in the drainage water compared to the unplanted one. They suggested that the difference in NO₃–N removals might be related to plant uptake processes since NO₃–N is the most favorable N form assimilated by plants. In the study of Fillion et al. (2009), willows and poplars growths reduced NO₃–N leaching even when high irrigation rates were applied. Indeed, in their work only samples collected from lysimeters installed in the control area where no trees had been planted showed the presence of NO₃–N in the drainage water.

Recent research on VFs focuses on the use of fast-growing tree species (Fig. 3), such as willows, poplars and eucalypts, with high water requirements and root systems tolerant to anaerobic conditions, which enables the application of large volumes of wastewater (Fillion et al., 2009; Herschbach et al., 2005; Persson and Lindroth, 1994). Other types of vegetation, such as grasses, crops or herbs, are also used, showing good performances as well (Barton et al., 2005; Duan and Fedler, 2010; Handley and Ekern, 1984; Kadam et al., 2009; Li et al., 2012, 2015; Ouazzani et al., 1996; Tzanakakis et al., 2007; Woodard et al., 2002; Zhang et al., 2005).

An important part of the water treatment capacity is related to the amount of water that the plants can evapotranspirate. Hence, the vegetation species is an important factor to consider when designing a VF (Fillion et al., 2009). Considering the most commonly used tree species, poplar and willows, the evapotranspiration capacity depends mainly on the availability of nutrients. Indeed, higher evapotranspiration rates are achieved when there is a higher proportion of available nutrients for the plant (Curneen and Gill, 2014; Pistocchi et al., 2009). Thus, nutrient supply must be evaluated along with the selection of the plant species to maximize nutrient uptake because of evapotranspiration processes.

The planting densities in VFs of the studies published between the 1990 and 2000 ranged from approximately 500 trees per hectare to 2000 trees per hectare (de Bustamante, 1990; Myers et al., 1995; Rigueiro-Rodriguez et al., 2010). However to allow better treatment performances and higher biomass production rates, much higher planting densities are used nowadays (generally between 10,000 and 20, 000 trees/ha) (Jarecki et al., 2012; Lazdina et al., 2007; de Miguel et al., 2014); except in cases where planting densities were already given (natural forests) or where researchers decided to use lower planting densities for operability purposes (Hashemi, 2013; Sanz et al., 2014). In some cases, densities of 30,000 trees/ha (Curneen and Gill, 2015) and 70,000 trees/ha (Białowiec et al., 2007) are reported (Białowiec et al., 2007; Curneen and Gill, 2015). Finally when the researches have been carried out in small pots, the equivalent planting density increases to the unrealistic value of 100,000 trees/ha (Bhati and Singh, 2003). In short rotation coppice (SRC) plantations, Holm and Heinsoo (2013b) concluded that the best density for the five more productive clones of willows, irrigated with municipal wastewater, would be 14,800 plants per hectare. On the other hand, Godley et al. (2005) compared the results in terms of yield of two SRC plantation in Hatfield (England) with densities of 12,000 and 24,000 trees/ha without finding any significant difference in terms of treatment performance. Since density plantation conditions the accessibility of certain machinery necessary for maintenance practices, this aspect has to be considered in the VF design.

As already described, the most used tree species in these systems are willows and poplars. One of the main reasons of their success is the tolerance they show to flooding (Fillion et al., 2009). The use of the genus *Salix* spp. is more propitious and suited for temperate-cold climates, such as those in northern Europe, while the genus *Populus* spp. is more suitable for temperate and Mediterranean climates, such as those in southern Europe (Dimitriou and Rosenqvist, 2011; de Miguel et al., 2014; Romano et al., 2013). This might be at least partially due to the fact that, under SRC conditions, willows show minor preference for NO₃–N over NH₄–N, compared to poplars, that prefer NO₃–N, according to the studies carried out by Sommer et al. (2017) using ¹⁵N labelling. Thus, this makes willows a better option for cold, acid and low

oxygenated soils than poplars.

Willows show a better response to fertilization than poplars. Guidi et al. (2008) found that the crop coefficient of willows and poplars increased by 56% and 38%, respectively, when the plantation was irrigated with nutrient-rich influents. Indeed, willows exhibit a greater evapotranspiration than poplars. This is consistent with results of Pistocchi et al. (2009) who found that, under optimum irrigation conditions, fertilization had a more pronounced influence in willows than in poplars. Pandey et al. (2011) also obtained higher increases in biomass production when fertilizing willows (by 275%) compared to poplars (by 55% in dry biomass production). However, in this case the total biomass production was greater for poplars (22,075 kg ha⁻¹ against 9450 kg ha⁻¹) since the study was carried out under the subtropical climate conditions of India.

As highlighted by Dimitriou and Aronsson (2011), to achieve the best removal rate of contaminants, wastewater irrigation should take place only when plants are well-established, otherwise extensive leaching can be expected. In this study, the authors also found that willows showed better performances than poplars during their first rotation cycle, as it took longer for the latter to develop a proper roots system.

Apart from the studies carried out with poplars and/or willows, other researchers compared the efficiency in the treatment with different plant species. Tzanakakis et al. (2007) evaluated soil NO3-N concentrations when using (besides Populus nigra) Eucalyptus camaldulensis, Acacia cyanophylla and Arundo donax to treat domestic wastewater under the same conditions. The highest NO3-N concentrations in soil were observed with Eucalyptus camaldulensis whereas the opposite occurred with Arundo donax. Similar results were obtained by the same authors few years later when they researched exclusively on Eucalyptus camaldulensis and Arundo donax (Tzanakakis et al., 2011). They suggested that the development of a larger underground biomass by Arundo donax may have contributed to the uptake of NO3-N, while the prevalence of conditions which promoted higher denitrification rates may have influenced as well. These two statements are, actually, very closely related, as a higher root development leads to better support for nitrifying organisms, as mentioned above (Perttu and Kowalik, 1997).

On the other hand, Woodard et al. (2002) compared the effectiveness for systems planted with i) Bermuda grass and ryegrass, and ii) corn, forage sorghum and ryegrass, under the same conditions. Bermuda grass removed more N from the soil than the corn-forage sorghum combination and, thus, the concentration of NO₃–N in leached water was lower for Bermuda grass than for corn-forage sorghum combination (6.3 mg/L versus 30.3 mg/L). The perennial characteristic of Bermuda grass vs annual forages as sorghum can be the reason of its better efficacy to prevent groundwater contamination by NO₃–N.

4.2. Soil

Soil is the most important compartment for wastewater treatment through VFs. It acts as a physical, chemical, and biological filter, and together with the roots of the plants is the support for microorganisms that are involved in the removal of contaminants. The elimination of nutrients in amended soils with an adequate proportion of OM (high proportion of labile and easily assimilable carbon) can reach values higher than 99% in column tests (Meffe et al., 2016) without the need of vegetation.

From a physical point of view, the texture and structure of the soil determine its porosity, which in turns conditions the infiltration rates. The elimination of N and P, among others, is highly dependent on the residence time of the water in the soil, as it influences the degree of contact between the soil colloids (clay and OM) and the components of the wastewater. The sorption of nutrients (N and P) and OM, onto soil mineral and organic constituents is one of the chemical processes controlling their fate, transport, and (bio)availability. Soil particles retain contaminants present in wastewater via different mechanisms (e.g. cation exchange, partitioning, surface complexation) depending on the

nature of the substance (Hillel and Hatfield, 2004). NH_4^+ -N is usually retained in soil by cation exchange processes. Texture and OM content, together with the pH, have a considerable influence on cation exchange capacity (CEC). This process is favored by small grain size textures, high pH and high OM content (Catalán Lafuente, 1997; Duchaufour, 1975) and it is highly dependent on the nature of the fine mineral (clays, oxides) and organic (functional groups) fractions. On the other hand, it can also occur that some nutrients in wastewater precipitate in contact with other species present in the soil, as it occurs with PO₄–P in calcareous soils (Bouwer, 1974; Duchaufour, 1984).

Nutrient removal depends also on the biological activity. Most of the wastewater treatment occurs in the upper soil horizons (<1.2 m depth), where a biologically active layer exists (Catalán Lafuente, 1997; Duan and Fedler, 2010). This zone is where the majority of roots are developed (Holm and Heinsoo, 2013b; Kotowska et al., 2009) and where the microbiota lives. One of the most important ways to eliminate N in a VF is via denitrification driven by this microbiota under the appropriate conditions (Duan et al., 2015; Hooda et al., 2003; Kadam et al., 2009; Li et al., 2012; de Miguel et al., 2014; Schipper et al., 1989; Tyrrel et al., 2002; Zhang et al., 2015). A high content of OM produces an increase in the microbial population and activity in such a way that denitrification is favored (Carrey et al., 2014; de Miguel et al., 2013). In more evolved soils with a higher OM content, the elimination of nutrients, and in particular of N, is enhanced. Thus, during the operation of a VF, as time elapses an OM-rich horizon is being created, and a more complex root system is being established. Therefore the leaching of N compounds is likely to be decreased (Aryal and Reinhold, 2015; Dimitriou & Aronsson, 2005, 2011; Guidi Nissim et al., 2015; Holm and Heinsoo, 2013b; Hooda et al., 2003; de Miguel et al., 2014; Rastas Amofah et al., 2012). This means that the performance of VFs in terms of contaminant attenuation should be evaluated considering a span of time long enough to let the system be fully developed. For instance, Dimitriou and Aronsson (2011) found strongly different treatment capacities comparing 3-year-old willows and 1-year-old poplars, but this differences disappeared in the second irrigation season, when poplars' treatment capacity increased due to its development. As a rule of thumb, the more the VF "gets older" the more its attenuation capacity increases. This is mainly due to the formation of an OM-rich horizon and the establishment of a complex root system (rhizosphere) that hosts and stimulates microbial activities (Aryal and Reinhold, 2015; Dimitriou & Aronsson, 2005, 2011; Guidi Nissim et al., 2015; Holm and Heinsoo, 2013b; Hooda et al., 2003; de Miguel et al., 2014; Rastas Amofah et al., 2012).

Coarse soils with high drainage and low OM contents have a poor denitrification capacity while sandy loam and loam soils have intermediate capacity. Higher degree of denitrification are usually obtained with silt and clay soils (Barton et al., 2005; Catalán Lafuente, 1997; Hooda et al., 2003). There are two main reasons behind this effect. On one hand, smaller particle sizes and clay minerals provide larger specific surfaces (Carter et al., 1986; Cerato and Lutenegger, 2002; Ersahin et al., 2006), which increases the CEC (Bayat et al., 2015). The higher CEC and surface areas in clay minerals can raise respiration rates of bacteria present in the soil, as clays with higher CEC are able to maintain adequate pH conditions for microbial respiration (Stotzky, 1966). On the other hand, fine textured soils are less permeable than those with coarse textures, which means higher residence times, higher microbial activity (Meffe et al., 2016) and therefore a better pollutant removal from the wastewater (Fillion et al., 2009). However, low N removals can occasionally occur also in fine textured soils as a result of preferential flows and poor root aeration (Barton et al., 2005; Dimitriou and Aronsson, 2011). In natural systems such as FVs, the selection of the soil is not always a possibility and experiences with coarse textures have been reported (Aggarwal and Goyal, 2009; Bhati and Singh, 2003; Farahat and Linderholm, 2015). In these cases, the success strictly depends on the improvement of the systems in terms of OM content and quality, specific surface areas and infiltration capacities as above mentioned.

4.3. Hydraulic load and irrigation systems

The irrigation system and schedule represent an additional factor that is crucial for the proper functioning of VFs. An inadequate irrigation operation, whether due to deficit, excess or misdistribution, is one of the main causes of failure. When designing a VF, it must be considered that they are natural systems with a vegetation having certain water requirements. There are different design and dimensioning methodologies for FVs. Based on our experience and given that nutrient concentrations in the influent are usually very variable, the most appropriate method to avoid water stress problems, is the water balance method (Cozzi et al., 2015; de Bustamante et al., 2009; Duan and Fedler, 2009; Zhou et al., 2006) rather than the nutrient load one (Aronsson et al., 2010; Cozzi et al., 2015; de Bustamante et al., 2009; Duan and Fedler, 2009; Zhou et al., 2006). Additionally, the incorporation of amendments that increase microbial activity and nutrients consumption could palliate a potential exceed of nutrients (Meffe et al., 2016) when the water balance method is applied.

Once the VF is designed and established, methods for systematic irrigation are various. Not all of them are always applicable, and they show different efficiencies in terms of plant development and nutrient removal. Irrigation systems range from sprinklers for secondary treatment effluents (Duan and Fedler, 2010; Edraki et al., 2004) and landfill leachates (Aronsson et al., 2010) to flood irrigation, either by blanket irrigation throughout the whole system (de Bustamante, 1990; Farahat and Linderholm, 2015; Tanvir and Siddiqui, 2010) or by furrows (de Miguel et al., 2014). Another method widely used is the drip irrigation (Aronsson and Bergström, 2001; Dimitriou and Aronsson, 2004; Guidi Nissim et al., 2014; de Miguel et al., 2014; Pistocchi et al., 2009; Truu et al., 2009), in some cases with self-compensating pipes (de Miguel et al., 2013), allowing for a more homogeneous distribution throughout the entire VF surface (Sanz et al., 2014). Perforated pipe without dripper is also used (Holm and Heinsoo, 2013a; Sanz et al., 2014). With sufficient water availability, this would be a good choice for achieving a homogeneous distribution. The advantages of using perforated pipes relies on: i) their easy compensation for pressure losses along the pipeline; ii) the minimization of the problems related to obstruction, often presented by drippers, and iii) their low cost.

In cold climates, and during the winter months, vegetation water needs are satisfied by the precipitation and therefore irrigation is not necessary (Kotowska et al., 2009). Sometimes and depending on the local climate, the water can be stored in rafts for its use during the dry season (Dimitriou and Aronsson, 2005). However, safely storage of wastewater is anything but straightforward (Cirelli et al., 2008; King, 1982; Yang et al., 2019). In addition to the water requirements of the plant, it must be considered that the recovery of nutrients also depends on its growth stage and on the temperature. Therefore, it is advisable to reduce or even eliminate irrigation during the winter months, depending on the attenuation capacity of the soil (Guo and Sims, 2000).

In general, as the hydraulic and contaminant loads increase, the system efficiency tends to decrease. In this sense, Li et al. (2012) recommended hydraulic loads of 81 L/m²·d and contaminant loads of 16.80 g BOD/m² for their systems in Liaoning province (China). Khan and Irvine (2011) also found that a reduction in the N load led to better performances in LASs treating meat industry effluents in New Zealand compared to historical performances with higher N loads. An equilibrium between the hydraulic and contaminant loads needs to be achieved for a satisfactory biomass production and effective wastewater treatment. In northern Europe climates, Perttu and Kowalik (1997) found that irrigation loads of 50 mm/week during the growing season would imply acceptable contaminant concentrations, with a satisfactory biomass production. In terms of biomass production, Hasselgren (1998) observed that willows irrigated with approximately 140 mm/week of wastewater during their growing period produced a biomass three times higher than those that were tap water irrigated and non-fertilized. After that period, the author found that loads of 42 mm/week seemed to be

optimal for growth while loads higher than 56 mm/week resulted in smaller biomass production. Differences in hydraulic loads between authors may partially respond to changes in soil characteristics. In the study of Hasselgren (1998), soils had a finer texture compared to those of the study carried out by Perttu & Kowalik (1997), enhancing residence times and allowing the application of more water (see Soil effects). However, sometimes an increase of hydraulic retention times can result in a decrease of TN removal related to the absence of anoxic conditions or, more likely, to the low quantity of organic matter available to denitrifiers (Collivignarelli et al., 2020).

Not only hydraulic loads but also irrigation patterns condition the treatment efficiency. In Li et al. (2011), the variation of treatment performance was investigated under intermittent operation modes with different wetting-drying ratios and continuous operation mode. Obtained results demonstrated that the intermittent operation mode significantly enhanced contaminant removal. Decreasing of wetting-drying ratios (from 1.0 to 0.25) had no effect on BOD, COD or TP concentrations. Whereas, NH₄-N removal rose, indicating that nitrification processes were favored by the improvement of the oxidative conditions during the drying period. However, transformation of NH₄-N to NO₃–N without further denitrification does not have a real impact on TN and this is the reason why Li et al. (2011) recommended a wetting-drying ratio of 1.0. for an optimum TN removal. This is consistent with what de Miguel et al. (2014) found when flooding each furrow one day a week, as the achievement of anaerobic-aerobic conditions allowed to stimulate nitrification-denitrification processes.

The selection of hydraulic loads also depends on climate conditions and specific problems of the region. For example, Ou et al. (1997) reported that in arid zones hydraulic loads should be high enough to guarantee the leaching of salts beyond the root zone avoiding secondary salinization of the land. These hydraulic loads were still safe enough to ensure that there were no further leaching and subsequent groundwater pollution. On the contrary, in countries with high precipitation rates, the hydraulic loads need to be lowered to maintain effective retention times.

In conclusion, it is clear that higher hydraulic loads lead to a reduction in contaminant attenuation, increasing the risk of ground-water contamination as a result of undesirable substances leaching (e.g. N). Nevertheless, high hydraulic loads also have other advantages, as they enhance biomass production and prevent salinization in dry areas. Therefore, the ideal hydraulic loads for each system must be assessed and may change depending on the season and site climate conditions. Additionally as proved by Li et al. (2015), effective hydraulic loads may be achieved with intermittent operation, and not only by reducing the amount of irrigation.

4.4. Nitrogen load

An excess of N loads when operating a VF may determine the leaching of NO₃–N towards the underlying aquifer. In the reviewed literature, the N loads applied are variable, ranging from 40 kg N/ha-year) (Labrecque et al., 1995) to over 300 kg N/ha-year (Adegbidi and Briggs, 2003; Dimitriou and Aronsson, 2004). Occasionally, the load exceeds 2000 kg N/ha-year in water from landfills in Sweden (Aronsson et al., 2010). The N loads recommended for the correct development of willow SRCs are usually between 60 and 170 kg N/ha-year (Dimitriou et al., 2012; Hansson et al., 1999; Hasselgren, 1998). Within these low N load range, the leaching of NO₃–N into groundwater is unlikely to occur (Cogliastro et al., 2001). This holds true especially for N loads smaller than 100 kg N/ha-year. However, Dimitriou & Aronsson (2011) showed that once the VF is well-established and has developed a proper root system, a nutrient load of 370 kg N/ha-year can be applied without compromising the safety of the underlying aquifer.

As already commented, one of the methods of VF design and dimensioning is based on the N balance. The purpose of this approach is to ensure that N concentrations of the water treated by the VF do not exceed at any moment the limits established by local administrations. The N balance method is meant to achieve this by matching N contents in the wastewater with plant requirements. However, the biggest flaws of this methodology are that calculated hydraulic loads do not fulfill the water requirements of the plants (de Bustamante et al., 2009) and that it ignores the N-attenuation performed by the microbiota, only considering plant uptake. A further limitation is represented by the natural variability of N concentration in the applied wastewater that hampers an efficient operation of the systems designed using the N balance approach (Dimitriou and Aronsson, 2011).

4.5. Type of wastewater

Studies about the performance of VFs report the use of many different types of wastewater, including urban, industrial and synthetic wastewater. Most of the papers reviewed described the use of municipal or domestic wastewater (Barton et al., 2005; Dimitriou and Rosenqvist, 2011; Duan et al., 2010; Holm and Heinsoo, 2013b; Li et al., 2012; Pan et al., 2016; Perttu and Kowalik, 1997; Tzanakakis et al., 2009). Nevertheless, there are also several studies considering industrial wastewater, rich in trace metals (Antil et al., 2001; Bhati and Singh, 2003; Rattan et al., 2005) or in nutrients and OM originated from manure or food industry (Aryal and Reinhold, 2015; Cavanagh et al., 2011; Christen et al., 2010; Khan and Irvine, 2011).

Concerning wastewater pretreatments, a great diversity can be observed. Most of the studies that used municipal and domestic wastewater applied primary or secondary treated wastewater (Barton et al., 2005; Duan et al., 2010; Li et al., 2011; Tzanakakis et al., 2009), reclaimed water (Chen et al., 2013) or even raw wastewater (de Bustamante, 1990; Li et al., 2012; Zhang et al., 2005) to their systems. In general, the use of secondary effluents vs primary effluents shows better removals of N but roughly the same results in terms of P attenuation. In Curneen and Gill (2016), systems irrigated with secondary wastewater showed a better performance for TN, especially for organic N, than those where primary treated wastewater were applied. Hasselgren (1998) reported that plants irrigated with secondary effluent produced slightly more biomass than plants irrigated with primary effluent, due to the higher fraction of NO₃-N in secondary wastewater. To our judgement, this is mostly related to the preferential plant uptake of NO3-N rather than NH₄-N (Barker and Mills, 1980; Cao and Tibbitts, 1993; Salsac et al., 1987). In the case of SRC, poplars tend to uptake more NO₃–N than NH₄-N, while willows uptake similar quantities of each N species (see 4.1 Vegetation) (Sommer et al., 2017). Other authors (Cao and Tibbitts, 1993; Carr et al., 2020) have demonstrated better plant growth when both N species are available. In addition, the presence of NO₃-N in secondary wastewater seems to facilitate N attenuation. Indeed, when VFs are irrigated with primary wastewater N removal depends on both nitrification and denitrification effective rates.

Concerning industrial wastewater, King (1982) compared the treatment efficiencies between pretreated and untreated industrial wastewater originated from a fiberboard mill compared to control plots (inorganic fertilizer). The authors found that pretreatment increased the amount of N present in pretreated wastewater compared to untreated wastewater, which lead to an increase in soil N when irrigated with treated wastewater, while soil irrigated with untreated wastewater showed similar NO₃-N concentrations as the control plots. Therefore, they suggest that plant uptake, the accumulation of N in the organic crust that was gradually formed, and nitrification-denitrification cycles were responsible for N attenuation, so there were no significant risks of pollutant leaching (N, P, K, Ca) when untreated wastewater was used. Nevertheless, they used only soil samples to evaluate the VF performance and stressed on the necessity of additional research to test the applicability of VF to treat different types of industrial wastewater. Indeed, industrial wastewater contains a great variety of contaminants that can be detrimental for the treatment capacity of the system. An example is presented by Antil et al. (2001) that found that the application of metal-rich industrial wastewater led to the increase in metal

soil contents affecting negatively the elimination of nutrients.

Concerning VF and more in general LAS application for food industry wastewater, there are several studies that provided promising results (Aryal and Reinhold, 2015; Christen et al., 2010; de Bustamante, 1990; Khan and Irvine, 2011). For example, wine industry originated wastewater enhanced C/N ratios and, therefore, N attenuation (de Bustamante, 1990). LASs seem to be a suitable strategy for the treatment of food industry wastewater, as long as good practices are implemented, instead of using it as a "dispose and forget" system (Khan and Irvine, 2011).

5. Effects on groundwater

Contaminant and hydraulic loads, irrigation patterns, soil, plant growing stage, plant coppice and time for VF stabilization are crucial factors to protect groundwater resources from contamination. One of the most important problems with VF's operation is the threat of groundwater pollution due to contaminant leaching. When water volumes applied to the VF exceed the potential evapotranspiration value and the water retained by the soil (i.e. plant water reserve), recharge of the underlying aquifer occurs. Under this scenario, the NO_3 -N that is not assimilated by plants or transformed by microbes is practically not retained by the soil and leaches towards groundwater.

In the literature, there are studies in which the application of liquid manure or domestic wastewater contaminated the underlying aquifer (Mantovi et al., 2006; Tzanakakis et al., 2007). In Mantovi et al. (2006), this impact was especially important in the presence of a bare soil. In Tzanakakis et al. (2007), the aquifer contamination increased with time due to nitrification rates exceeding the plant NO_3 –N uptake capacity. Another example in which the operation of VFs resulted in groundwater pollution is described by Aronsson et al. (2010), using a willow plantation to treat landfill leachate. In this study, the levels of N and trace metals in groundwater indicated a massive overload of the system. In the research reported by Guidi Nissim et al. (2015), NO₃–N concentrations in groundwater rose without exceeding legal values. These examples show that it is necessary to balance application loads with the N plant uptake and adjust it to their growing stage, as exposed in Hasselgren (1998).

Duan et al. (2010) found that potential groundwater pollution by N is minimized when wastewater irrigation follows a schedule based on the water balance method specifically thought for the experimental site (Texas, USA) and described in Duan and Fedler (2009). In the study carried out by de Miguel et al. (2014), there were no significant differences in NO₃-N concentrations in groundwater before and after the 3-year operation of the VF. Only during the first year when the plantation was in its early growth stage groundwater concentrations showed slightly higher values than background levels. These results are similar to those obtained by Dimitriou and Aronsson (2011), who concluded that the amounts applied to their SRC (370 kg N/ha/year and approx. 30 kg P/ha/year) would not contaminate groundwater thanks to the efficiency of the VF in contaminant attenuation when plantations are well established. Same conclusions were achieved in the study of Aronsson and Bergström (2001), where wastewater irrigation of willow SRC has no substantial risk of NO3-N pollution in groundwater. Cameron et al. (1995) applied pig slurry to increase pasture production in low amounts (200 kg N/ha·year) without posing a serious threat to groundwater. However, these results changed when N loads were raised to 600 kg N/ha-year.

6. Effective improvements of VFs

Once they are well-stablished, plants in their early stages of growth have much higher water and nutrient requirements. For this reason, these systems have evolved from long-term plantations to obtain wood of high quality, with rotation periods of 12–15 years (De Bustamante, 1990; Rigueiro-Rodriguez et al., 2010), to plantations for biomass

production with shorter rotation periods, between 2 and 4 years (Aronsson et al., 2010, 2014, 2014; Aronsson and Bergström, 2001; Sean Curneen and Gill, 2015; Dimitriou and Aronsson, 2005; Hooda et al., 2003; de Miguel et al., 2014). Thus, once trees begin to resprout, the ability to evapotranspirate water and absorb nutrients will be greater than having continued with normal growth (Dimitriou and Aronsson, 2005; Tzanakakis et al., 2009). Among others, denitrification is also favored when plantations are coppiced, due to the higher insulation and subsequent higher temperature in soil (Hooda et al., 2003). According to Guidi Nissim et al. (2013), after three or four cuts, this practices can considerably improve VF performance. However, it is not really clear if what affirmed by Guidi Nissim et al. (2013) is valid for every case. After several rotation, yield capacity and biomass production for both willows and poplars depends on many factors, including mortality after coppicing, re-sprouting capacity (both related to the selected genotype), local climate conditions, soil quality and duration of each rotation (2-3 years growing periods show better yields than yearlong growing periods) (Harayama et al., 2020; Paris et al., 2011; Pérez García, 2016).

Primary treatment application such as settlers (pond systems or septic tanks) tends to enhance the system performances, preventing groundwater contamination (Christen et al., 2010; Duan and Fedler, 2010; Li et al., 2015; Tzanakakis et al., 2011). An outstanding efficacy of settlers is observed especially for wastewater suspended solids for which removals above 60% can be achieved. On the other hand, the implementation of settlers has a lower impact on N and P removal (less than 10%) and COD and BOD removal (aprox. 20%) (Li et al., 2015). Different configuration of a settler like Imhoff tanks, causes a suspended solids reduction of 50-70%, COD reduction of 25-50%, and leads to potentially good sludge stabilization - depending on the design and conditions (Stauffer and Spuhler, 2019). Occasionally, primary treatment methods include the addition of several amendments as adsorbent (coke powder, lignite powder, active carbon, pulverized coal and black soil) and coagulants (aluminium chloride, alum, animal glues, marine algae, freshwater algae, fibers and pectic substances) (Zhou et al., 2006).

Zhou et al. (2006) improved the VF by designing and developing secondary plant covers as back-up systems to face the problems due to the low or even inexistent removals during winter periods. These secondary plant covers consisted in smaller (10-15% of VF area) and ecologically diversified areas (weeping willows, chinese scholartrees, magnolias, horsebeans and rice) which received high hydraulic loads especially during the vegetative growth arrest of rice and crops forming the main land treatment system. A similar strategy was suggested by Mantovi et al. (2006), and used by Torstensson and Aronsson (2000), where cover crops were planted in early autumn and were harvested in the following spring. This represents an interesting way of facing a very common problem, when the plants have lower activities and soil denitrification is the most important N removal process (Martinez, 1997). An additional possibility suggested by Mantovi et al. (2006) and Tzanakakis et al. (2007) is to adjust the N application loads during winter (or during the preceding months) to the capacity of the system to assimilate and remove this nutrient, always considering the type of soil.

A very common strategy to improve VF performances is the application of irrigation schedules with drying periods. Wetting-drying cycles promote nitrification-denitrification processes, due to changes from oxidizing to anoxic conditions in the soil, and therefore enhances N removal in the system. Soil aeration is crucial since nitrification is often the limiting factor for N removal in VFs treating raw or primary treated wastewater, due to the shortage of oxygen (Kuusemets et al., 2001). In Zhang et al. (2005), the application of an intermittent operation mode (24 h flooding period followed by 24 h of drying period) doubled the soil nitrification potential and increased NH₄–N removals from 70 to 90%. These results agree with those obtained by Li et al. (2015), who also found that the NH₄–N removals increased as the drying period was prolonged. As reported by Duan et al. (2015), a further strategy that can be implemented to prevent groundwater pollution is the installation of subsurface drainages. These systems collect the effluent (or leaching water) and divert them to another cropped system, using it as a fertilizer.

Finally, the addition of soil amendments should receive the attention they deserve. In general terms, amendments such as woodchips, biochar or sewage sludge, among others, can improve VF performances by increasing OM content, sorption surface area, CEC, aggregation, porosity and water holding capacity of the soil, factors that are characteristic of highly evolved soils (Arval and Reinhold, 2015; Dimitriou & Aronsson, 2005, 2011; Guidi Nissim et al., 2015; Holm and Heinsoo, 2013b; Hooda et al., 2003; de Miguel et al., 2014; Rastas Amofah et al., 2012; Singh and Agrawal, 2008). The improvement of these soil characteristics provides in turn beneficial impacts by mainly promoting microbial activity and sorption capacity (Martínez-Hernández et al., 2020; Meffe et al., 2016). In this sense, Martínez-Hernández et al. (2020) described how the addition of poplar woodchips to soil was responsible of enhancing microbial activity while biochar soil addition fostered sorption processes. Overall, woodchips clearly provided the best N removal percentages, while biochar did not have a comparable impact. One of the aspects that should be emphasized when using woodchips or biochar is that these amendments can be obtained from maintenance pruning of forests or gardens, transforming a waste in a resource, or directly from the VF pruning, as an example of circular economy.

The application of sewage sludge as an amendment was studied and several works concluded that it enhances VF biomass production, while it did not result in higher leaching (Dimitriou and Aronsson, 2011; Dimitriou and Rosenqvist, 2011; Holm and Heinsoo, 2013a; Kalisz et al., 2012; Kocik et al., 2007; Kotowska et al., 2009; Lazdina et al., 2007; Moffat et al., 2001; Rigueiro-Rodriguez et al., 2010). This is consistent with the observations that Singh and Agrawal (2008) reported in their review about land application of sewage sludge. The authors claimed that it improves soil properties such as porosity, bulk density and water holding capacity while the problems derived from its use only arises when it contains high contents of trace metals, as their availability can cause toxicity problems to the plants and affect the enzymatic activity in the soil. Indeed, the application of sewage sludge in agriculture and the concerns about its application to the land date back to the 70's of the 20th century (Singh and Agrawal, 2008), and the introduction of trace metals is a matter of concern that has been considered in the Directive 86/278/EEC on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture.

7. Outlook on future research

To our judge, there are plenty of research aspects about VFs that can be still tackled. For example, more research is needed to evaluate the use of these systems in different scenarios involving different types of water (i. e. industrial wastewater -including metal rich wastewater- and stormwater runoff). In this sense, as already mentioned, the use of soil amendments is a wide field of study that requires dedicated efforts for material selection and configuration depending on water and soil characteristics. Indeed, many natural materials, in most cases considered as waste, can be useful to achieve higher yields in wastewater treatment.

Another interesting issue to be investigated is the plant selection according to the applied wastewater. VFs are very similar to SRCs, and there are plenty of different genotypes that can be used in this type of forestry plantations. Despite being the same species, these genotypes, present different behaviors, production rates and resilience to different conditions. Thus, it is very likely that some of these genotypes show better response than others depending on wastewater characteristics and/or climate conditions.

Also, there is a lot of interest in ecological benefits that VFs provide, and future research should be driven to quantify these benefits through standardized evaluation methods allowing for comparison among NBSs. These environmental benefits are especially important in lowbiodiversity areas, such as industrial sites or agricultural fields (Vanbeveren and Ceulemans, 2019).

8. Conclusions

This review provided data about the performance of VFs to treat wastewater, elucidating the role of parameters that are considered by the authors as crucial for a proper operation. VFs have multiple benefits that render this technology a suitable solution especially for scattered populations or isolated buildings that lack of connection to sewer systems. However, the operation easiness of these systems somehow incites the end-users to have a "dispose and forget" attitude, a problem that can be tackled by implementing a simple but rigorous operation and maintenance schedule to avoid system failure.

Several works have reported solid results about VF performance and main conclusions are:

- VFs are found to be a suitable system for wastewater treatment, achieving good removals of N, P, BOD, COD. Nevertheless, they require a simple but rigorous operation and maintenance schedule to avoid the system failure.
- Leaching of contaminants in VFs are mainly related to too high hydraulic and contaminant loads, and coarse sand soils with too low retention times and sorption capacity, together with physicochemical conditions affecting the microbial community in the rhizosphere. However, high hydraulic loads also have advantages, as they enhance biomass production and prevent salinization in dry areas. Therefore, ideal hydraulic loads for each system must be assessed and may change depending on the season and site climate conditions.
- Willows and poplars show great biomass production potential and uptake capacity. Plant growing stage, plant coppice practices, VF development and stabilization and suitable wastewater application rates are crucial factors for a correct implementation of the system and, therefore, for the protection groundwater resources.
- The water balance method seems to be the most valid method to assess wastewater application rates, as it fulfils plant water requirements and facilitates the planning of the irrigation schedule according to the season and the activity of the plants. On the other hand, when N concentrations in applied wastewater are high, the nutrient load method is likely to provide insufficient water for plant growth.
- Besides some exceptions, primary and secondary treatment systems allow the achievement of better removals comparted to the application of raw wastewater. This holds true especially for TSS, COD and BOD.
- Application of amendments, including sewage sludge with low trace metal concentrations, promotes microbial activity, and ameliorates soil properties, leading to nutrient removal improvements. In this sense, these simple practices could help to minimize leaching risks when the nutrients load is higher than plant uptake capacity.

Credit authorship contribution statemen

Raúl Pradana: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Visualization. Jorge Hernandez-Martín: Conceptualization, Methodology, Formal analysis, Data curation, Writing – original draft. Virtudes Martínez-Hernandez: Conceptualization, Methodology, Writing – review & editing, Supervision. Raffaella Meffe: Conceptualization, Methodology, Writing – review & editing, Supervision. Ana de Santiago-Martín: Conceptualization, Methodology, Writing – review & editing, Supervision. Adrián Pérez-Barbón: Methodology, Formal analysis. Irene de Bustamante: Conceptualization, Funding acquisition, Writing – review & editing, Supervision. Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial

interests or personal relationships that could have appeared to influence the work reported in this paper.

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R. Pradana et al.

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R. Pradana et al.

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