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Research article

# Soil amendments and water management to improve attenuation and recovery of wastewater originated nutrients through a vegetation filter

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# ABSTRACT

Vegetation filters (VFs) are on-site wastewater (WW) treatments that can be considered as a nature-based solution (NbS). They are green infrastructures that provide several environmental benefits such as non-potable water reuse, contamination reduction, biomass production, landscaping improvements and  $CO_2$  fixation, among others. However, nutrient leaching, especially nitrate, partially exists. To overcome this limitation, operational parameters related to the irrigation water management and soil amendments were tested in a real system receiving WW from an office building operated along 4 years.

The attenuation of N is improved (up to 83%) in the vadose zone by boosting biodegradation. Lower hydraulic loads and more frequent irrigation events using drippers and the incorporation of woodchips as a layer above the topsoil promote denitrification processes. Changes in organic carbon characteristics also confirm that biodegradation is enhanced.

P attenuation is a result of abiotic processes, mainly driven by chemical equilibriums between the liquid and the sorbed and/or precipitated phase and, when uncontrolled changes in the WW quality occurs, removal efficiency is negatively affected. However, only 10% of the samples collected at 45 cm depth present concentrations above 2 mg/L. The woodchips application does not seem to ameliorate P removal regardless of the application method.

The implemented measures allow higher soil water content, infiltration and groundwater recharge and prevents aquifer contamination.

# 1. Introduction

Vegetation Filters (VFs) are on-site treatments considered as a type of Nature-Based Solutions (NbSs) that benefit from natural processes occurring in soil ecosystems to treat and reclaim urban wastewater in low densely populated areas where technical and economical limitations hamper the effective implementation of conventional treatments. VFs share with other NbSs the great potential to face the actual water management challenges such as water pollution and deteriorating water quality (EEA, 2021; UN-Water, 2018). Moreover, in the context of a sustainable urban development, their applicability as a measure to maintain and/or enhance ecosystem services contributes to tackle climate change (Sutherland et al., 2014).

A VF is a plant-based treatment where pretreated wastewater (WW) rich in nitrogen (N), phosphorus (P) and organic matter is applied to a plantation, usually formed by tree species, under a controlled setting. The contaminants present in the WW undergo attenuation processes during infiltration through the soil as a result of the joint action of sorption, biodegradation and phytoabsorption. Poplars are the species most commonly used in VFs due to their ability to develop the root system predominantly by lateral extension down to a 40–50 cm depth (Douglas et al., 2016; Jerbi et al., 2015), concentrating the active rhizome (biological treatment) in the uppermost soil horizon. They are also fast-growing species providing interesting benefits in biomass production especially when managed through short rotation coppice (SRC).

The capacity and efficiency of removing nutrients in VFs has been largely recognized (Pradana et al., 2021). However, there are still some

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Abbrev	riations	GW	groundwater
		GWr	groundwater recharge
NbS	Nature based Solutions	Pz <sub>up</sub>	upgradient piezometer
VF	vegetation filter	Pzdown	downgradient piezometer
SRC	short rotation coppice;	Т	temperature
WW	wastewater	Р	precipitation
WWv	wastewater irrigation applied volume	$K_c$	crop coefficient
HLR	hydraulic loading rate	ETO	evapotranspiration
INW	infiltration water	RET	real evapotranspiration
θ	volumetric water content in soil	TN	total nitrogen
$\theta_0$	water content in soil before irrigation	KTN	Kjeldahl total nitrogen
$\theta_{s}$	water content at saturation conditions	TP	total phosphorous
$\theta_{FC}$	volumetric water at field capacity	DOC	dissolved organic carbon
ST	soil saturation time;	SUVA <sub>254</sub>	specific ultraviolet absorbance
DT	soil drainage time;	TOC	total organic carbon

limitations in their treatment efficiency causing nutrient leaching during infiltration, particularly of nitrate (de Miguel et al., 2014; Lasa et al., 2011). This holds true especially in those cases in which heterogeneous and high irrigation loads are applied (Al-Jamal et al., 2002; Vázquez et al., 2006). A homogeneous surface distribution of water promotes the effective development of the root surface improving water uptake while reducing infiltration rates (Camposeo and Rubino, 2003; Du et al., 2018; Segal et al., 2006). The irrigation regime controls the water residence time in the rhizosphere which is crucial for minimising contaminant leaching (Dukes et al., 2010; Lasa et al., 2011; Li et al., 2015; Zotarelli et al., 2011).

Natural attenuation in VFs can be further improved through the addition of soil amendments (Pradana et al., 2021) that can also enhance biomass production (Miller and Seastedt, 2009). Some waste organic by-products with a sorbent capacity higher than soil potentially increase retention processes when they are used as amendments (Ahmad et al., 2014; De Gisi et al., 2016). However, they are not always effective to avoid nutrient leaching (Lu et al., 2022; Morgan et al., 2020) or provide ambiguous results when their performance is evaluated taking into account all the different species of N (Shrestha et al., 2019; Yin et al., 2017). Due to their composition, carbon-rich substrates are excellent candidates as soil amendments, particularly those derived from agricultural products such as woodchips, pellets, and peat, that has proven highly effective in reducing the leaching of contaminants and nutrients, such as N and P (Chen et al., 2014; Meffe et al., 2016; Pradana et al., 2021; Robertson, 2010). Their basic components include hemicelluloses, lignin, lipids, proteins, simple sugars, hydrocarbons and starch (Bhatnagar and Sillanpää, 2010) containing a variety of functional groups with a potential sorption capacity for various contaminants (Bhatnagar et al., 2015; De Gisi et al., 2016). Most of them also serve as a source of organic matter that stimulates microbial degradation (Becagli et al., 2021; Soria et al., 2021). Among carbon-rich substrates, the efficiency of woodchips as soil amendments to promote microbial activity and, therefore attenuate nutrients has been already demonstrated (Martínez-Hernández et al., 2020; Meffe et al., 2016). They enhance soil moisture retention, thereby mitigating leaching and evaporation of retained water, which in turn promotes root development and indirectly supports contaminant attenuation by microorganisms and plants. Additionally, the surface of woodchips can act as a sorbent for inorganic compounds such as ammonium ions (NH<sub>4</sub><sup>+</sup>), thereby reducing the concentration of dissolved contaminants in infiltrating water (Wang et al., 2014). Moreover, results from our most recent study (Martínez-Hernández et al., 2020) indicate that woodchips are even more effective in coping with N leaching compared to biochar, a material with a well-known sorption capacity. Both studies were performed under controlled conditions at laboratory scale and upscaling woodchip incorporation into soil under environmental conditions is therefore

crucial to consolidate obtained results.

The main objective of this study is to maximize the treatment efficiency of N and P from WW using a VF. To this end, a pilot VF receiving WW from an office building was operated under environmental conditions and monitored for approximately four years. Different operational parameters (frequency of irrigation, HLR and irrigation system) as well as soil amendment configurations were tested to unfold their impact in terms of nutrient treatment efficiencies.

# 2. Material and methods

## 2.1. Site description and VF design

The pilot VF (40°30′48.6″N 3°20′15.4″W), managed through SRC of poplars, was designed to treat the WW generated at the IMDEA Water (Alcalá de Henares, Spain). The pilot VF (48 m2) consists of a high-density plantation (10,000 plants/ha) of Populus euroamericana (clone I-214) with a planting pattern of  $1 \times 1$  m (Fig. 1) using the ridge-furrow method.

Before its application, the WW is pretreated by a 9 m<sup>3</sup> Imhoff tank whose effluent is directly discharged into a pumping well from where it is distributed to the pilot VF. To size the VF, the water balance (Eq. (1)) was inferred considering the inflows, or wastewater irrigation applied volume (WWv) and natural precipitation in the area (P), the outflows as the plantation water requirements or evapotranspiration (ET), the volume of infiltrated water based on the soil field capacity ( $\theta_{FC}$ ) and soil water storage (R). The water needs of the plants are calculated daily to estimate the amount of irrigation water that must be applied to fulfil the plant water requirements or exceed this value to foster infiltration for groundwater recharge (GWr) purposes.

Water Balance =  $(WWv + P) - (ET_0 \cdot k_c) \pm (\theta_{FC} - R) > 0 \rightarrow GW_r$  (1)

The water needs of the plants are calculated daily based on climatic data obtained from the Spanish Agroclimatic Information System (SIAR) and the plant growth stage. The information was retrieved from the nearest agroclimatic station *CENTER Finca el Encín* in San Fernando de Henares (Madrid, Spain). A continental Mediterranean climate, with an average annual rainfall of 489 mm and average annual temperature (T) of 14.3 °C (AEMET, 2022) is characteristic of the study site. The temperature ranges between 0 and 18 °C from November to April and between 5 and 33 °C from May to October.

To estimate the daily evapotranspiration of poplars and of the herbaceous plants that spontaneously grow in the VF, the corresponding crop coefficients (Kc) (Allen et al., 2006) were multiplied by the reference evapotranspiration (ET<sub>0</sub>) and resulting values were summed to account for both vegetative species (Table A.1). Plant water needs were occasionally not fulfilled as a consequence of the limited WW production

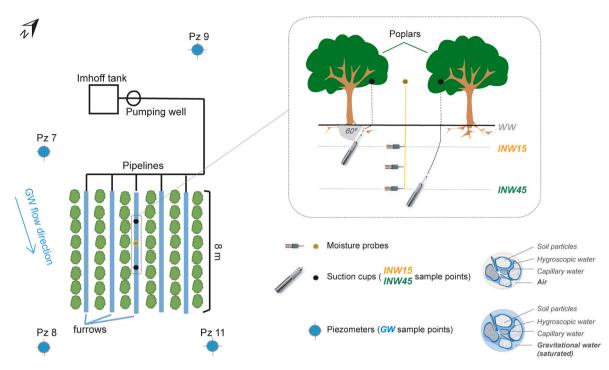


Fig. 1. Scheme of the pilot VF with the location of sampling points (suction cups and piezometers) and monitoring equipment (moisture probes). The drawing is not to scale.

at IMDEA Water during holiday periods and during the teleworking regime established as a sanitary containment measure against the COVID-19 pandemic.

#### 2.2. Experimental setup

Given the objective of maximizing WW treatment, and consequently nutrient uptake, a SRC based on 2-year pruning was chosen as the appropriate cultivation technique. Part of the wood material resulting from the pruning was further processed to obtain the woodchips used for soil amendments. This study reports a 4-year research devoted to maximizing contaminant removal by optimising operational parameters. During the operation of the VF, three periods have been differentiated (Table 1) according to: i) the irrigation method and regime (in terms of frequency and HLR), and ii) the addition of amendments to the VF soil (42.5% sand, 26% silt and 31.5% clay).

In the first (P1) (January 2018–April 2019) and second (P2) (April 2019–July 2020) period, WW was applied by flooding each furrow once per week (5 working days per week).

The P2 begins with the soil amendment after the first pruning of the VF. Hence, 3% (w/w) woodchips were mixed with the uppermost 15 cm of the soil upscaling the promising results obtained in our previous research at laboratory scale (Martínez-Hernández et al., 2020). Finally in period 3 (P3) (July 2020–October 2021), a 10 cm woodchip layer was added over the irrigation furrow surface, the drip irrigation system replaced the furrow irrigation, and WW was applied on a daily basis.

#### 2.3. Sampling and measurements

To assess the VF treatment performance, samples were retrieved from the WW, infiltrating water (INW) and groundwater (GW) following the flow path during infiltration. Fig. 1 shows the sampling points and the equipment installed in the VF along with its location.

Shortly before reaching the VF, the WW volumes were measured by a flowmeter installed in the main pipe and whose data were retrieved at least once per day. The main pipe was then ramified into five sub-pipes distributing the water to the corresponding furrows of the VF. WW samples were collected from the sub-pipe located in the middle of the pilot where the equipment used for monitoring the infiltration dynamic and INW quality was installed to avoid edge effects.

The INW was collected using two single chamber stainless steel suction cups of 260 mL, installed at 15 and 45 cm depth and connected to the surface by polytetrafluoroethylene tubes. Prior to their installation, the devices were rinsed according to standard procedures recommended by providers and installed at a distance of 2 m between them. The vacuum was applied to the stainless steel cups through a hand pressure pump equipped with a manometer or through an automatic vacuum pump (VS-Pro) operating between 3.5 and 4 bars.

The study of the infiltration dynamic provides complementary information to interpret data about INW quality. For this reason, the volumetric water content ( $\theta$  - m3/m3) was monitored by means of three moisture probes (ECH2O-5 TE) installed at 15, 30 and 45 cm depth. For the analysis on how attenuation processes may be influenced by the way water circulates in the unsaturated zone, data obtained by the moisture probes were graphically analysed. The  $\theta$  temporal variations describing the infiltration dynamic, mainly depends on soil characteristics and applied irrigation volumes (WWv), and it was investigated by selecting  $\theta$ values in winter when evapotranspiration is negligible. To this end, three crucial points were considered in each weekly temporal moisture curve: water content before irrigation ( $\theta$ 0), at saturation ( $\theta$ s) and at field capacity ( $\theta_{FC}$ ) (Zotarelli et al., 2010). Average water contents along with standard errors were calculated for each crucial point and operational period (P1, P2 and P3). To determine the duration of the gravitational drainage through the macropores, saturation and drainage time intervals corresponding to select water content values were calculated. Specifically, saturation time (ST) encompasses all intervals during which the water content was above mean  $\theta_{\text{s}}$  minus its standard error and drainage time (DT) is the sum of the intervals during which water contents are above mean  $\theta_{FC}$  plus its standard error (Table A.4 and Fig. A.1). Four piezometers distributed in the surrounding area of the VF (Fig. 1) were used to monitor the groundwater table oscillation, whereas the groundwater quality was controlled exclusively using two of them. Two piezometers, one upgradient (Pzup) and one downgradient (Pzdown) (10 m depth) of the experimental plot (Pz 9 and Pz11, respectively)

#### Table 1

Management of described periods along with changes in operational parameters, soil amendments and sampling and measurements schemes in the pilot VF.

		Period	P1	P2	Р3
			January 2018 - April 2019	April 2019 - July 2020	July 2020 - October 2021
Management		Soil amendments	-	woodchips mixed with topsoil	woodchip layer over the topsoil
		Irrigation method Irrigation	furrow irrigation weekly	furrow irrigation weekly	drip irrigation daily
Monitoring	Water	frequency In-situ basic parameters	monthly	weekly	daily
		•			
		In-situ nitrate		weekly	weekly
		Major ions		weekly	weekly <sup>a</sup>
		•			
		TN, TP, COD, TOC, DOC <sup>b</sup>		monthly	monthly
		-			
		SUVA <sup>b</sup>	с	с	monthly
		-			
		-			
	Soil	Moisture, pH, EC TN, NO <sub>3</sub> -N, TP, organic matter CEC <sup>b</sup> and major ions <sup>d</sup>	yearly	yearly	yearly

<sup>a</sup> Composite sample from merged aliquots retrieved daily. ■ Wastewater (WW), ■ Infiltrating water (INW), ■ Groundwater (GW).

<sup>b</sup> Analized parameters (Total Nitrogen – TN; Nitrate - NO<sub>3</sub>-N; Total Phosphorous – TP; Chemical Oxygen Demand – COD; Total Organic Carbon – TOC; Dissolved Organic Carbon - DOC; Specific Ultraviolet Absorbance - SUVA<sub>254</sub>; Cation Exchange Capacity – CEC).

<sup>c</sup> SUVA254 analyzes began in the mid-P3. For earlier periods (P1 and P2), data are only available for sample aliquots stored at that time.

 $^d$  Major ions (Cl<sup>-</sup>, NO<sub>2</sub>, NO<sub>3</sub>, HPO<sub>4</sub><sup>2-</sup>, SO<sub>4</sub><sup>2-</sup>, CO<sub>3</sub><sup>2-</sup>, HCO<sub>3</sub>, OH<sup>-</sup>, Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup>)

(Fig. 1) were indeed periodically sampled and analysed to evaluate potential contaminant leaching from the VF.

To recognize changes in soil properties and to obtain additional data supporting result interpretation, soil has been collected down to a depth of 15 cm at the beginning of the study, for an initial characterization, and once per year during the three periods (four soil sampling campaigns in total). To account for spatial heterogeneity, soil samples were retrieved from a minimum of six locations evenly distributed in the VF using a manual auger. A composite sample was then obtained by quartering the soil directly at the pilot site. Prior to analysis, samples were air-dried, gently crushed and passed through a 2-mm sieve.

For the sake of clarity, the information concerning the timing of sampling and analyses for each experimental period (P1, P2 and P3) has been structured in Table 1 differentiating among *in-situ* basic parameters (pH, electrical conductivity - EC, redox potential and dissolved  $O_2 - DO$ ), *in-situ* nitrate (NO<sub>3</sub><sup>-</sup> in GW), major ions (Cl<sup>-</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, HPO<sub>4</sub><sup>2-</sup>, SO<sub>4</sub><sup>2-</sup>, CO<sub>3</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup>, OH<sup>-</sup>, Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup>), nutrient (Total Nitrogen - TN and Total Phosphorous - TP) and organic matter (Chemical Oxygen Demand – COD, Total Organic Carbon - TOC and Dissolved Organic Carbon – DOC and Specific Ultraviolet Absorbance - SUVA<sub>254</sub>).

#### 2.4. Parameters and laboratory analyses

The in-situ basic parameters (pH, EC and redox potential) were measured directly in the field with a multimeter MM-41 (Crison, Spain). The in-situ  $NO_3^-$  measurements in GW were carried out through ultraviolet spectrophotometry using a portable multiparametric analyzer PASTEL-UV (Surcis, Spain).

Major ions were analysed using a 930 Compact Ion Chromatography Flex (autosampler 858 Professional Sample Processor) coupled to a Titrando 809 (autosampler 814 USB Sample Processor) for  $HCO_3^-$  determinations. Analyses of TN, TP, TOC and DOC (Table A.2) were performed by spectrophotometry according to standardised methods reported in (Eaton et al., 2005).

The UV absorbance of water samples was performed by visible-UV spectrophotometer (UV-1800 Shimadzu) at a wavelength,  $\lambda$  of 254 nm (UV254). SUVA254 is then calculated dividing the UV254 by sample DOC content (Cornu et al., 2011). This parameter provides a general characterization of the organic matter nature indicating the degree of its aromaticity and, therefore, indirectly providing information about its recalcitrance (Weishaar et al., 2003).

Soil EC and pH were determined in a 1:5 ratio of soil:water. Soil organic matter content was measured by the Loss-On-Ignition method at 360 °C for 24h. Soil nitrate content was analysed by ion chromatography after extraction as described by (Griffin et al., 2011). The analysis of the Kjeldahl Total Nitrogen (KTN) was carried out following the standardised method UNE 77318:2001. TP was determined using ICP-MS, after microwave-assisted acid digestion and assimilable phosphorous by Olsen method (Olsen et al., 1954). The cation exchange capacity (CEC) was determined by extraction with ammonium and sodium acetate solutions. After soil extraction, Na + concentration was analysed using ICP-MS.

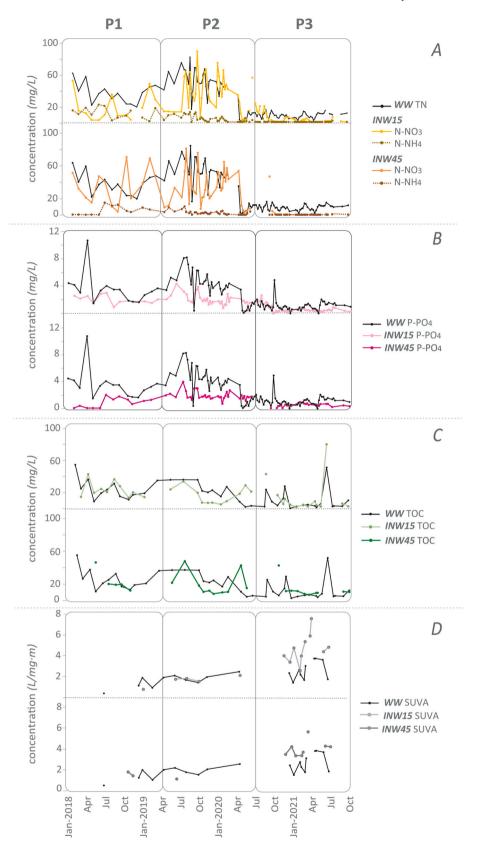
#### 3. Results and discussion

#### 3.1. Concentration evolution and attenuation in the unsaturated zone

The evolution of N and P species (TN, N-NO<sub>2</sub>, N-NO<sub>3</sub>, N-NH<sub>4</sub>, and P-PO<sub>4</sub>) and organic matter concentration in WW and INW for both monitoring depths (15 and 45 cm) during the three study periods (P1, P2 and P3) is presented in Fig. 2. The corresponding attenuation values (%), calculated weekly by comparing the concentrations in WW with those measured in INW at the two depths and GW, are shown in Fig. 3. In the case of N, attenuation percentages were calculated in terms of TN provided by spectrophotometric analyses. Whereas P attenuation was evaluated taking into account exclusively P-PO4, considered to be the prevalent species in WW and INW. The organic matter attenuation (%) were calculated using TOC data. In the following sections, nutrient and organic matter attenuation processes will be discussed considering the impact of the irrigation management and amendment applications during the three study periods.

## 3.1.1. Nitrogen

TN concentrations in WW were variable and dependent on the working activity in the building. During P1 and P2, TN contents were quite similar (44.8  $\pm$  4.8 mg/L and 54.3  $\pm$  5.8 mg/L, respectively)



**Fig. 2.** Nutrient concentration in irrigation water (WW, 0 cm), vadose zone (INW, -15 and -45 cm depth) during the three study periods. (**A**) Total Nitrogen (TN) content (mg/L) in WW and N species (ammonia N-NH<sub>4</sub> and nitrate N-NO<sub>3</sub>) contents (mg/L) in INW. (**B**) Phosphate contents (P-PO<sub>4</sub>) (mg/L). (**C**) Total Organic Carbon (TOC) (mg/L) concentrations and (**D**) calculated SUVA<sub>254</sub> (L/mg·m).

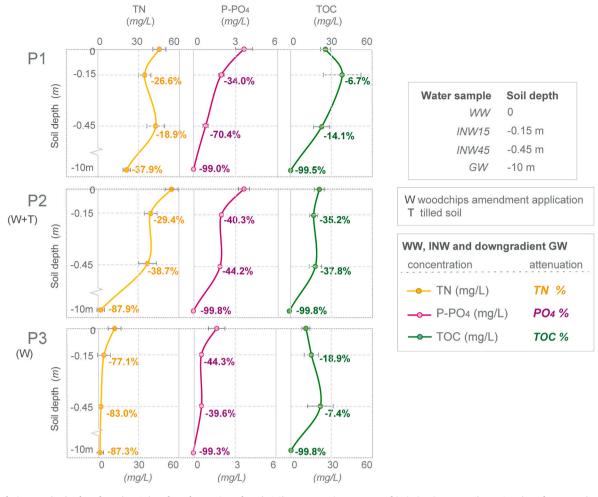


Fig. 3. Total nitrogen (TN), phosphate (P-PO<sub>4</sub>) and total organic carbon (TOC) concentrations measured in irrigation water (WW, 0 cm), vadose zone (INW, -15 and -45 cm depth) and downgradient groundwater (GW, -10 m) and their attenuation percentages during infiltration in the three study periods.

whereas during the last period, the limited presence of the personnel in the building due to the teleworking regime caused a drop in TN concentrations (11.8  $\pm$  0.9 mg/L) (Fig. 2A). This N is mainly in the N-NH<sub>4</sub> form (86.7  $\pm$  4.1%, 75.1  $\pm$  4.4% and 71.6  $\pm$  3.3% in P1, P2 y P3, respectively). As expected, N-NO<sub>3</sub> and N-NO<sub>2</sub> contents in WW were negligible (<1 mg/L), independently of the study periods (Table A.2).

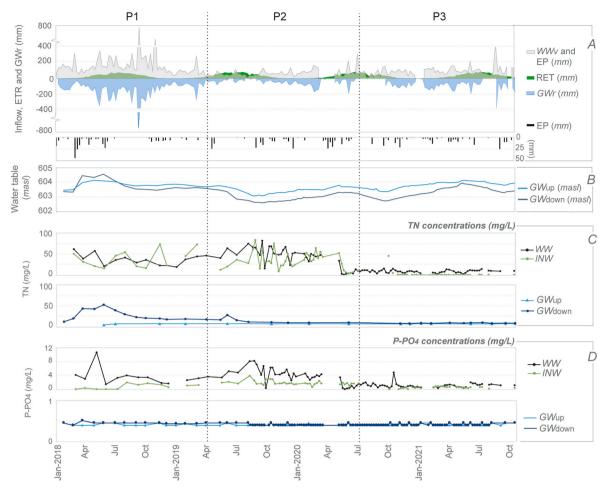
Variability among periods was also observed in the INW data (Fig. 2A). Likewise, the TN average concentrations in INW during P1 and P2 were again similar (33.9  $\pm$  4.5 mg/L and 38.9  $\pm$  5.8 mg/L at 15 cm depth; 42.1  $\pm$  6.6 mg/L and 36.6  $\pm$  5.4 mg/L at 45 cm depth). Whereas, during P3, the TN average concentration in the INW was much lower (3.9  $\pm$  1.2 mg/L and 1.7  $\pm$  0.4 mg/L at 15 and 45 cm depths) partially due to the reduced levels in the WW (Table A.2). Differences among periods were even more evident when analysing the speciation of N as a consequence of reactive processes taking place during infiltration. During P1, up to 45.9  $\pm$  7.9% of the TN leaching at 15 cm was in the form of N-NH<sub>4</sub>, whereas during P2 and P3 the main form of TN leaching corresponded to N-NO<sub>3</sub> (84.4  $\pm$  3.1% and 71.4  $\pm$  3.6%, respectively) indicating the occurrence of nitrification processes.

An effective approach to evaluate the efficiency of the pilot VF to attenuate WW originated nutrients was by comparing the attenuation percentages among the three periods. In this way, the effect of concentration variation related to factors out of our control (i.e. personnel presence in the building) is somehow called off. As a general evaluation, the applied measures in terms of irrigation regime, irrigation systems and soil amendments with woodchips resulted in the improvement of N attenuation, whose maximum values were reached during P3 at 45 cm depth (83.0  $\pm$  3.1%) (Fig. 3).

Differences in treatment efficiencies among the three periods can be explained by various causes. In P1, the limited percentages of TN attenuation (26.6  $\pm$  4.7% and 18.9  $\pm$  6.5% at 15 and 45 cm depth, respectively) (Fig. 3) correlates with the N-NH<sub>4</sub> leaching. At 45 cm depth, the N-NO<sub>3</sub> leaching occurred with 3-month delay with respect to 15 cm depth indicating the slow but progressive saturation of sorption sites (Fig. 2A). Such conditions are particularly fostered under continental or Mediterranean climates when large irrigation volumes are applied (Díaz et al., 2019; Vázquez et al., 2006). Therefore, the high HLR (31% of the irrigations exceed the water needs of the VF) (Fig. 4A) applied through furrow irrigation in P1 was the main responsible for N losses. In addition, the limited development of the microbial activity at the rhizosphere during the early-stage growth of trees implied that nutrient sorption onto soil mineral phases is likely to be the dominant attenuation phenomenon.

During P2, N-NH<sub>4</sub> leaching progressively decreases. Towards the end of this period, concentrations lower than the analytical detection limit (0.19 mg/L) were observed. The amendment incorporation and the topsoil remobilization by manual tillage fostered aeration conditions of the soil, promoting nitrification processes, increasing the permeability and, therefore, the infiltration rate through the uppermost 15 cm. The increase in macroporosity and permeability results in a higher  $\theta_{S}$  (0.369  $\pm$  0.002 m<sup>3</sup>/m<sup>3</sup>) (Table A.4 and Fig. A.1) if compared to that registered during P1 (0.331  $\pm$  0.001 m<sup>3</sup>/m<sup>3</sup>).

In addition, tillage labour in clay-loam soils favours the desorption of organic matter retained onto the clay surface (Oades, 2018). This now



**Fig. 4.** (A) Groundwater recharge (GWr) function of water volumes applied ((wastewater irrigation (WWv) and precipitation (EP)) and real evapotranspiration (RET). EP data (black bars) are presented separately given the difference in scale with applied irrigation (**B**) Upgradient ( $Pz_{up}$ ) and downgradient ( $Pz_{down}$ ) water table measurements. (**C**) TN (mg/L) and (**D**) P-PO<sub>4</sub> (mg/L) concentrations measured in WW, INW (at 45 cm depth),  $GW_{up}$  and  $GW_{down}$ . Note that the smaller range of values of the y-axis is used for a better visualization of GW quality data.

available organic matter was likely added to that provided by the amendments, contributing to raising the C/N ratio of the soil (Table 2). The increase of the organic carbon under oxidizing conditions promoted the growth of nitrifying bacterial populations (Van Veen and Kuikman, 1990). The aspects hitherto discussed explain the fluctuation in N-NO<sub>3</sub> concentrations in the INW, the reduced N-NH<sub>4</sub> leachate and the still limited TN removal.

Such reduced treatment performance of the VF continued temporarily during the transition from P2 to P3 (April–September 2020). The low N concentrations in the WW enhanced soil desorption of previously accumulated N. However, leaching of N to deeper levels was not observed because of reduced HLRs that prevent water infiltration. The absence of water infiltration was also corroborated by data from soil moisture probes ( $\theta_S$ ) and such a behaviour lasted throughout P3. Under these circumstances, it is evident that N moved through the soil profile, but it remained confined within the root zone.

Finally, the highest removal percentages of TN were obtained during P3 (77.1  $\pm$  4.7% and 89.4  $\pm$  3.1% at 15 and 45 cm, respectively) (Fig. 3). The improvement of the system efficiency was given by the convergence of the following factors: (i) the new irrigation management through more frequent irrigation using drippers, (ii) the amendment application without altering the soil structure and (iii) the change in the WW quality that implied a reduced N concentration due to the teleworking regime. The drip irrigation method as an alternative to furrow

Та	ıble 2				
-		C . 1			

Evolution of the soil physico-chemical	l parameters during the study perio	ods.
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Parameter	Feb 2017 (start)	Mar 2019 (P1)	$\Delta$ start-P1 (%)	Jul 2020 (P2)	∆ P1-P2 (%)	Mar 2021 (P3)	∆ P2-P3 (%)
рН	8.2	8.1	-1.6	8.1	+0.2	8.4	+2.7
CE $(dS/cm)$	140.0	170.0	+25.7	212.0	+20.5	144.0	-32.1
Organic Matter (%)	1.7	1.8	+7.7	2.9	+61.0	3.0	+1.7
CEC (mg/kg)	11.1	15.9	+43.2	13.9	-12.6	15.2	+9.4
KTN (mg/kg)	760.0	950.0	+25.0	1290.0	+35.8	1330.0	+3.1
$NO_3^-$ (mg/kg)	45.5	125.0	+174.7	130.0	+4.0	26.4	-79.7
TP $(mg/kg)$	394.0	404.3	+2.6	457.0	+13.0	409.0	-10.5
Assimilable P (mg/kg)	8.0	27.6	+245.0	28.4	+2.9	32.4	14.1
TOC (%)	1.0	1.1	+7.7	1.7	+61.0	1.7	+1.7
KTN (%)	0.1	0.1	+25.0	0.1	+35.8	0.1	+3.1
C/N	12.9	11.1	-13.8	13.2	+18.6	13.0	-1.4

irrigation reduces the nitrogen leaching fraction (Pool et al., 2022) and to provides a more homogeneous water distribution (Zotarelli et al., 2011). Indeed, changes in the application mode of the WW and the reduced HLR accompanied by higher irrigation frequency were all together responsible for achieving the treatment goals. The treated WW volume was not reduced in this last period since weekly treated volumes were similar to those of the previous period ( $70.55 \pm 6.23$  mm and  $95.77 \pm 7.4$  mm in P2 and P3) and the lower HRLs were compensated by a sharp increase of the irrigation frequency. This increase caused more recurrent wetting-drying cycles which in turn influence the simultaneous occurrence of nitrification-denitrification processes (Duan and Fedler, 2007) reducing N leaching (Amiot et al., 2020; Dukes et al., 2010; Lasa et al., 2011; Zhang et al., 2012)

The new woodchip layer applied as an amendment on the topsoil surface contributed to keep the high C/N ratio achieved through the first amendment in P2 (Table 2) without altering the infiltration dynamic at the rhizosphere. As suggested by Aalto et al. (2022), the lignocellulose contained in the woodchips promotes the growth of arbuscular mycorrhizal fungi that further improves the bioavailability of carbon for denitrifying bacterial communities (Zhai et al., 2021) and the transfer of N to the plant (Fillion et al., 2011). However, a well-structured rhizosphere is essential to maintain the cohesion among mineral particles and plant water retention, fostering the development of the soil microbiome (Dimitriou and Aronsson, 2011), which may have failed in P2 as a consequence of soil remobilization. Finally concerning changes in WW quality, the nutrient attenuation improvement and leachate reduction correlated with lower nutrient loads, similarly to what has been observed by other authors (Abu-Zreig et al., 2003; Ren et al., 2010)

Measurements of soil physico-chemical parameters (Table 2) supported previously discussed results. Their difference at the end of P1, P2 and P3 with respect to the immediately previous period, is also presented. At the end of P1, there was an increase in both KTN and NO<sub>3</sub> (+25.0% and +174.7%, respectively). However, in P2 despite the KTN concentration rose in an even higher proportion (+35.8%), the NO<sub>3</sub> only rose by 4%. In P1, the sorption processes promoted by the high N concentrations in the WW were responsible of both increases (NH<sup>4</sup><sub>4</sub> and NO<sub>3</sub>). In P2, NO<sub>3</sub> sorption capacity was likely reached and leaching of this species occurred. The outstanding reduction of soil NO<sub>3</sub> contents (-79.7%) observed in soil samples from the campaign of March 2021 (P3) and the absence of relevant N-NO<sub>3</sub> leaching concentration at 15 cm depth strongly suggest the occurrence of denitrification along with plant uptake.

#### 3.1.2. Phosphorus

Natural attenuation processes occurring in the VF approximately halved P-PO<sub>4</sub> concentrations contained in the WW. More specifically, average P-PO<sub>4</sub> attenuation percentages were 41.2  $\pm$  3.0% and 46.4  $\pm$  3.4% at 15 and 45 cm, respectively (data calculated for the entire study period), with some exceptions related to uncontrolled changes in the WW quality that alter the treatment efficiency during the experiment.

The main P-PO<sub>4</sub> attenuation mechanisms were adsorption processes onto positively charged surfaces present in soil (Dimitriou and Aronsson, 2011; , Tunesi et al., 1999) and P-PO<sub>4</sub> precipitation in the presence of  $Ca^{2+}$  (Eriksson et al., 2016) which was highly abundant in the VF soil (4952.5 ± 338.6 mg/kg). The TP content increase in the uppermost soil layer (15 cm) during P1 (+2.6%) and P2 (+13.0%) (Table 2) suggests that these processes were taking place. P attenuation during infiltration through the soil of VFs has been previously observed (de Miguel et al., 2014; Meffe et al., 2016) and other authors also found P sorption and precipitation during prolonged wastewater irrigation (Dimitriou and Aronsson, 2011; Lachapelle-T. et al., 2019) that, moreover, is likely accentuated in clay and calcic soils (Morel et al., 1989). The prolonged use of WW for irrigation can cause saturation of sorption sites (Brzezińska et al., 2011) and concentration variations can affect the solubilization of immobile nutrients in the mineral phase such as P (Tzanakakis et al., 2011). Indeed, the decrease in the removals at 45 cm depth in the first part of P2 (from  $70.4 \pm 6.9\%$  in P1 to  $44.2 \pm 4.5\%$  in P2) (Fig. 3) could be related to the increase of the concentrations in the WW. Sorption and precipitation are abiotic processes driven by geochemical equilibriums which are highly dependent on the WW chemistry. Indeed, changes in P-PO<sub>4</sub> loads and concentrations affected levels of this species in the INW and soil. In P3, leaching of previously sorbed P-PO<sub>4</sub> was likely fostered by nutrient imbalance between concentrations in the WW and contents in soil (-10.5% of TP content with respect to P2).

Moreover, during the transition from P2 to P3 (April–September 2020) (Fig. 2B), the teleworking regime caused low loads of P-PO<sub>4</sub> in the WW that altered chemical equilibrium in soil reducing P-PO<sub>4</sub> attenuation. These fluctuations in the WW chemistry caused the desorption of immobile P (Sande et al., 2005), especially in the presence of pH drops (Garrido Valero, 1994), such as those measured in the WW during P2 and transition to P3 (-0.7 from P1 to P2-P3). Also, the woodchip addition increased the humic acid content in the soil. These compounds can compete with P for sorption sites (Daly et al., 2001; Varela et al., 2017))favouring further P-PO<sub>4</sub> release to the aqueous phase and lowering the attenuation.

Following the transition period, an improvement on P-PO<sub>4</sub> removal in P3 (September 2020–October 2021) was observed at 15 cm depth (Fig. 2B), suggesting that sorption and precipitation were again effective after the major lixiviation. In previous periods (P1, P2 and P2-P3), the removal at 45 cm was higher than at 15 cm demonstrating that a larger unsaturated zone favored abiotic processes. However, in P3, removal percentages at 15 cm were higher than at 45 cm (44.3  $\pm$  4.7% and 39.6  $\pm$  5.7%, respectively) (Fig. 3). The reasons for this behaviour could be either related to fostered microbial activity in the upper soil horizon (Deubel and Merbach, 2005) or to ongoing lixiviation towards the lower soil horizon that counteracts sorption and precipitation processes occurring after the transition period. Nevertheless, over the entire study period, only 10% of data showed a P-PO4 concentration in the INW at 45 cm that is above 2 mg/L meaning that this species does not pose a risk concerning its subsurface transport towards groundwater.

#### 3.1.3. Organic matter

TOC contents in the WW followed a trend similar to that of the other nutrients presenting higher concentrations in P1 and P2 (25.6  $\pm$  3.6 and 21.8  $\pm$  3.7 mg/L) than in P3 (10.8  $\pm$  2.9 mg/L) (Fig. 2C and Table A.2). SUVA<sub>254</sub> was measured only in selected water samples to characterize its recalcitrant behaviour and data is provided in Fig. 2D and Table A.2.

TOC attenuation through the soil was low in P1 and P3 (<20%) while it reached an average value of 37.8  $\pm$  8.7% during P2 (Fig. 2C and Table A.2). Besides the incorporation of organic matter through the first amendment, the highest TOC attenuation in the INW during P2 coincided with the soil tillage, which favour the oxidation and easy biodegradation of the most labile compounds (Gursoy-Haksevenler and Arslan-Alaton, 2020). Woodchips can also provide recalcitrant compounds such as for example lignine (Kadjeski et al., 2020), but their contribution seems to be negligible since SUVA254 values during infiltration did not show differences from WW in P2. Similarly to what observed for N, biodegradation of organic matter was likely occurring due to the fostered microbial activity. During P3, SUVA<sub>254</sub> values indicated that biodegradation is also occurring despite TOC attenuation was rather low (20.4  $\pm$  5.1 %). In this case, removals were probably underestimated since calculations are biased by the fact that they only consider WW as the unique organic matter input instead of taking into account also the input provided by the second woodchip amendment.

### 3.2. Groundwater

The groundwater levels periodically monitored at both piezometers ( $Pz_{up}$  and  $Pz_{down}$ ) are represented in Fig. 4B. With the only exception observed in P1 (from March to July 2018), the groundwater level at the  $Pz_{down}$  was always below the level monitored at the upgradient piezometer confirming the general flow direction. From March to July 2018, the rise of the piezometric level in the  $Pz_{down}$  produced a local inversion of the flow direction. Such a behaviour likely correlates with the pronounced GWr obtained as a consequence of the heavy rain events (up to 43.9 mm per event) and the elevated HLRs applied to the VF (Fig. 4A). However throughout P1, the piezometric levels at both piezometers but specially at  $Pz_{down}$ , were higher than in the following periods (up to +0.8 m) suggesting the presence of a water source input from the surrounding area, outside IMDEA Water facilities.

During both P2 and P3, the trend of the groundwater level was very similar, with seasonally dependent oscillations. The levels at both piezometers declined during summer by 0.79 m in P2 and 0.48 m in P3 whereas during winter periods levels progressively recover. The hydraulic gradient (i.e.  $\Delta h/\Delta x$ , where h is the piezometric level and x the distance between piezometers) varied from a maximum of 0.95 % and 1.02 % to a minimum of 0.42% and 0.24 % in P2 and P3, respectively. The smaller hydraulic gradients observed in P3 were likely the results of the GWr achieved through the modified operating conditions (Fig. 4 and Section "3.3 Water balance").

Concerning chemical data, the GW in  $Pz_{down}$  presented an average pH of  $7.60\pm0.02$  and an EC of  $719.0\pm10.8\,\mu\text{S/cm}$ . The redox potential (154.0 $\pm$ 3.0 mV) and DO concentrations (5.3 $\pm$ 0.1 mg/L) are typical of an aquifer with moderate oxidizing conditions. The EC, pH, redox potential and DO remained practically invariant throughout the study period at both piezometers (Table A.3).

As expected, the dominant N species in groundwater was N-NO3 (Table A3). Average concentrations of N-NO2 and N-NH4 for each study period did not show any trend and presented levels lower than 0.06 and 0.24 mg/L, respectively. The outstanding rise of the water table described above and occurring from March to July 2018, was accompanied by an increase of TN, mainly in the form of N-NO3 (up to 52 mg/ L) in Pz<sub>down</sub>. Data about TN concentrations in Pz<sub>up</sub> were only available from May 23rd, 2018 however, despite being incomplete, they indicated that such a condition is circumscribed to downgradient GW. The rise in TN concentrations in the downgradient GW cannot only be explained by the leaching of this nutrient from the VF. As reported in Fig. 4C, TN levels in the water infiltrating at 45 cm depth (INW45) were similar or even below (from 18 to 63 mg/L) levels detected in GW. Even if TN in the form of N-NO<sub>3</sub> may infiltrate towards the aquifer with practically invariant concentrations, once in the groundwater it undergoes mechanical dispersion and dilution. Therefore, it is very likely that these transitorily high levels of N-NO3 in the aquifer were mainly due to an unknown contamination source from surrounding areas. From July 2018 onwards, TN concentrations progressively declined to 12.7 mg/L, but still above background levels (7.8 mg/L) until January 2020 when background levels were practically reached and steadily maintained throughout the rest of P3.

Independently of the study period, detected concentrations of P-PO<sub>4</sub> are smaller than 0.1 mg/L (the method detection limit). Concerning TOC, data reveal a progressive decrease from 2.14  $\pm$  0.29 mg/L (average concentration in P1) to < 1 mg/L (in P3) in the downgradient piezometer.

### 3.3. Water balance

The GWr by means of treated WW is an added value of VFs. The GW chemical analyses and the water balance calculations provided

information about the quality and quantity of the water recharge. GWr took place when the applied WWv exceeded the vegetation water needs and the soil water retention capacity ( $0.320 \pm 0.001 \text{ m}^3/\text{m}^3$  calculated at 45 cm depth during winter months). At the same time, GWr also depended on the irrigation method and regime (in terms of frequency and HLR).

The difference of GWr between P1 and P2 (weekly irrigations through furrows) mostly depended on the WWv applied. The greatest values calculated for P1 (136.3  $\pm$  13.6 mm/week vs 61.2  $\pm$  5.0 mm/ week) (Fig. 4A) corresponded to the WWv oversupply (159.08  $\pm$  14.76 mm) which was exacerbated by the limited water requirements during the early stage of poplar development (Jerbi et al., 2015).The decrease of GWr in P2 was also the result of the soil tillage for soil amendments. Indeed, the soil tillage increased macroporosity of the uppermost 15 cm layer (larger  $\theta_{\rm S}$ ; Table A.4), aeration, and therefore, ETP between irrigation events (lower  $\theta_0$ ; Fig. A.1 and Table A.4).

During P3, a constant soil water reserve was guaranteed promoting therefore GWr. Although the applied WWv resembles that of the previous period, the change to daily drip irrigation produced lower HLRs (27.3  $\pm$  2.7 mm vs 70.55  $\pm$  6.23 mm) responsible for longer residence times and a more permanent soil moisture. The values of DT during P3 double those calculated for P1 and P2 (30.5  $\pm$  3.6 h vs 13.3  $\pm$  2.0 h and 15.2  $\pm$  1.0 h; Table A4) confirming such a behaviour.

All data hitherto reported clearly indicate that the VF did not have a negative impact on the groundwater quality of the underlying aquifer. This holds true especially for P3 during which the drip irrigation and the applied regime ensured a slow but constant aquifer recharge preventing contamination.

#### 4. Conclusions

An appropriate management and combination of operational parameters related to water irrigation and soil amendments in VFs improve the attenuation of WW originated contaminants. The current work compiles and interprets a large quantity of chemical and physical analysis of soil and water matrices during a 4-year study to assess the processes involved in this attenuation. Such results allow us to provide solid conclusions at relevant environmental conditions that can be easily transferred to other VFs.

The attenuation of N is improved in the vadose zone (up to 83%) by lower HLRs and more frequent irrigation events with a homogeneous distribution of the water through drip irrigation. The incorporation of woodchips as a layer above the topsoil is preferred to amendments through soil tillage (woodchips mixed with soil). The woodchip applied as a top layer guarantees a carbon source that increases soil organic matter and augments the C/N ratio without altering the soil structure. All these conditions foster water retention and likely the presence of soil microbiome in the rhizosphere boosting biodegradation. Denitrification in soil is promoted by recurrent wetting-drying cycles, infiltration is reduced and residence times increased, rendering nutrients more available for plant uptake. Changes in organic carbon characteristics according to SUVA<sub>254</sub> measurements confirm that biodegradation is enhanced. It should be noted that the reduction of WW concentrations also contributed to improve treatment efficiencies.

P attenuation is a result of abiotic processes, mainly driven by chemical equilibriums between the liquid and the sorbed and/or precipitated phase and, when uncontrolled changes in the WW quality occurs, removal efficiency is negatively affected. Constant and low concentration loads assure a better removal efficiency of P. However, P-PO<sub>4</sub> does not represent a threat to the integrity of the groundwater since only 10% of the samples collected at 45 cm depth present concentrations above 2 mg/L. Regardless of the application method, woodchip amendments do not seem to ameliorate P removal.

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The application of excessive irrigation loads negatively affect the elimination of N and organic matter and may allow the propagation of these contaminants to downgradient groundwater. However, the implemented measures during P2 and, especially, P3 prevent groundwater contamination. During these last periods, the hydrochemistry of the groundwater is a consequence of natural background concentrations.

# CRediT authorship contribution statement

Lucía Barbero: Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. Virtudes Martínez-Hernández: Writing – review & editing, Supervision, Methodology, Investigation, Conceptualization. Blanca Huidobro-López: Writing – review & editing, Investigation, Formal analysis. Raffaella Meffe: Writing – review & editing, Supervision, Methodology, Investigation, Conceptualization. Irene de Bustamante: Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

### 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.

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## Appendices.

# Table A.1Crop coefficients (Kc) for poplars and grass (Allen et al., 2006).

	Jan.	Feb.	Mar.	Apr.	May.	Jun.	Jul.	Aug.	Sep.	Oct.	Nov.	Dec.
K <sub>c</sub> poplar	0.00	0.00	0.00	0.40	0.70	0.70	0.70	0.70	0.70	0.70	0.00	0.00
K <sub>c</sub> grass	0.24	0.38	0.55	0.85	0.90	0.95	1.00	1.00	0.95	0.80	0.54	0.35

Table A.2	
Physico-chemical measurements and nutrient concentrations in irrigation (WW) and infiltration water (INW	)

	P1			P2			Р3			
	ww	INW 15	INW 45	WW	INW 15	INW 45	WW	INW 15	INW 45	
pН	$8.20\pm0.11$	$8.11\pm0.06$	$\textbf{7.95} \pm \textbf{0.05}$	$\textbf{7.88} \pm \textbf{0.08}$	$8.02 \pm 0.05$	$\textbf{7.94} \pm \textbf{0.05}$	$\textbf{7.93} \pm \textbf{0.04}$	$\textbf{8.29} \pm \textbf{0.03}$	$8.18\pm0.05$	
EC (µS/cm)	$693.13\pm38.39$	$813 \pm 48.52$	1036.92 $\pm$	$747.15\pm37.3$	993.63 $\pm$	947.78 $\pm$	502.53 $\pm$	981.11 $\pm$	1219.46 $\pm$	
			56.03		38.75	39.46	22.93	50.43	126.36	
Redox (mV)	$-276.99~\pm$	75.99 $\pm$	$84.36\pm4.39$	$-217.86~\pm$	144.68 $\pm$	147.59 $\pm$	$88.49 \pm 6.96$	139.30 $\pm$	$124.89\pm5.55$	
	13.87	8.08		24.61	8.02	5.59		4.21		
DO *	$1.37\pm0.22$	$\textbf{8.86} \pm \textbf{0.74}$	$3.44 \pm 1.47$	$1.41\pm0.27$	$8.54\pm0.21$	$\textbf{8.24} \pm \textbf{0.42}$	$5.81 \pm 0.29$	$8.61\pm0.11$	$8.81\pm0.14$	
TN *	$44.83 \pm 4.82$	33.88 $\pm$	$42.10\pm 6.56$	$54.31 \pm 5.82$	$38.92 \pm 5.77$	$36.58 \pm 5.44$	$11.83\pm0.93$	$3.94 \pm 1.18$	$1.73\pm0.40$	
		4.89								
NH4-N *	$\textbf{37.04} \pm \textbf{3.53}$	11.54 $\pm$	$5.19 \pm 1.52$	$39.82 \pm 3.82$	$\textbf{4.09} \pm \textbf{0.64}$	$\textbf{2.46} \pm \textbf{0.38}$	$\textbf{8.08} \pm \textbf{0.47}$	$0.33\pm0.10$	$0.22\pm0.02$	
		1.93								
NO3-N *	$0.063\pm0.004$	$18.7\pm4.77$	$34.12\pm6.17$	$0.11\pm0.02$	$\textbf{35.46} \pm \textbf{3.81}$	$30.67\pm3.3$	$0.41\pm0.10$	$\textbf{3.75} \pm \textbf{0.84}$	$1.8\pm1.42$	
NO2.N*	$0.05\pm0.01$	$0.34\pm0.06$	$0.33\pm0.05$	$0.06\pm0.02$	$0.32\pm0.06$	$0.35\pm0.11$	$0.27\pm0.13$	$0.04\pm0.01$	$0.025\pm0.003$	
Att. TN (%)		$26.59~\pm$	$18.94 \pm 6.51$		$29.37 \pm 8.16$	$38.68 \pm 8.32$		$77.06 \pm 4.68$	$83.04 \pm 3.09$	
		4.68								
P-PO <sub>4</sub> *	$3.61\pm0.64$	$2.01\pm0.17$	$0.86\pm0.21$	$3.64\pm0.35$	$2.01\pm0.13$	$1.88\pm0.10$	$1.70\pm0.57$	$0.62\pm0.08$	$0.55\pm0.04$	
Att. P-PO <sub>4</sub>		$34\pm7.12$	$70.38 \pm 6.87$		$40.32 \pm 4.47$	$44.23 \pm 4.52$		$44.31 \pm 4.72$	$39.59 \pm 5.68$	
(%)										
TOC *	$25.56 \pm 3.62$	$37.89 \pm$	$22.78 \pm 5.94$	$21.81 \pm 3.66$	$17.67 \pm 2.9$	$18.93 \pm 4.51$	$10.85\pm2.93$	$14.75\pm5.29$	$21.3\pm9.37$	
		13.78								
DOC *	$11.21\pm2.11$	$23.77~\pm$	$19.30\pm3.80$	$12.37\pm3.08$	$\textbf{22.25} \pm \textbf{2.84}$	$31.6\pm0.0$	$3.48 \pm 0.35$	$6.82\pm0.68$	$9.81 \pm 0.79$	
		9.34								
SUVA***	$1.22\pm0.28$	<0.5	$1.64\pm0.19$	$1.78\pm0.29$	$1.93\pm0.12$	1.15 <sup>§</sup>	$\textbf{2.72} \pm \textbf{0.28}$	$\textbf{4.77} \pm \textbf{0.44}$	$\textbf{4.05} \pm \textbf{0.27}$	
Att. TOC (%)		$6.7\pm3.93$	$14.11 \pm 7.92$		$35.21 \pm 8.55$	$\textbf{37.76} \pm \textbf{8.67}$		$18.91 \pm 6.54$	$20.37 \pm 5.07$	

\*(mg/L). \*\*(cm $^{-1}$ ). \*\*\*(L/mg·M).  $\S$  (only one measurement was performed). Table A.3

Physico-chemical measurements and nutrient concentrations in groundwater upgradient ( $Pz_{up}$ ) and downgradient ( $Pz_{down}$ ) of the VF with corresponding standard error.

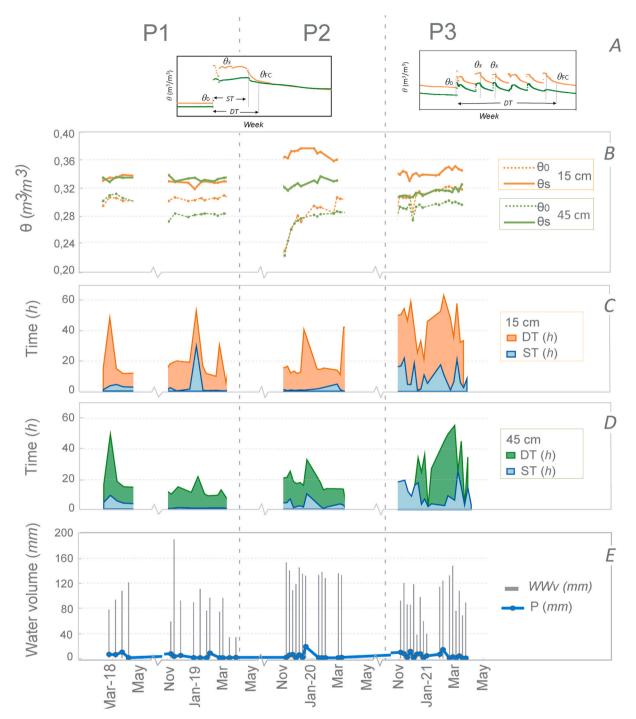
		рН	CE (µS/ cm)	DO (mg/ L)	Redox (mV)	TOC (mg/ L)	TN (mg/L)	N-NO <sub>2</sub> (mg/ L)	N-NO <sub>3</sub> (mg/ L)	N-NH4 (mg/ L)	TP (mg/L)	P-PO <sub>4</sub> (mg/ L)
P1	$Pz_{up}$	$\begin{array}{c} 8.00 \ \pm \\ 0.09 \end{array}$	$669 \pm 11$	$\textbf{4.4} \pm \textbf{0.4}$	$126\pm11$	$1.41 \pm 0.68$	$\begin{array}{c} \textbf{7.33} \pm \\ \textbf{0.68} \end{array}$	$\textbf{0.10} \pm \textbf{0.03}$	$\textbf{7.53} \pm \textbf{0.09}$	$\textbf{0.24} \pm \textbf{0.05}$	<0.03	<0.10
	Pzdown	$\begin{array}{c} \textbf{7.86} \pm \\ \textbf{0.06} \end{array}$	$785 \pm 15$	$\textbf{4.4} \pm \textbf{0.4}$	$157\pm10$	$\begin{array}{c}\textbf{2.14} \pm \\ \textbf{0.29}\end{array}$	$\begin{array}{c} 19.00 \pm \\ 1.10 \end{array}$	$\textbf{0.04} \pm \textbf{0.02}$	$\begin{array}{c} 14.75 \pm \\ 0.97 \end{array}$	$0.15\pm0.02$	$\begin{array}{c} \textbf{0.03} \pm \\ \textbf{0.01} \end{array}$	<0.10
P2	$Pz_{up}$	$\begin{array}{c} \textbf{7.97} \pm \\ \textbf{0.04} \end{array}$	$677\pm4$	$\textbf{5.8} \pm \textbf{0.8}$	$135\pm 6$	<1.00	$\begin{array}{c} \textbf{7.93} \pm \\ \textbf{0.09} \end{array}$	$\textbf{0.02} \pm \textbf{0.01}$	$\textbf{7.05} \pm \textbf{0.07}$	$\textbf{0.17} \pm \textbf{0.02}$	<0.03	<0.10
	Pz <sub>down</sub>	7.64 ± 0.04	$705\pm5$	$\textbf{5.0} \pm \textbf{0.1}$	$154\pm5$	$\begin{array}{c} \textbf{0.83} \pm \\ \textbf{0.21} \end{array}$	$\begin{array}{c} 14.48 \pm \\ 2.14 \end{array}$	$0.01\pm0.00$	$\textbf{9.68} \pm \textbf{0.18}$	$0.13\pm0.03$	$\begin{array}{c} 0.03 \pm \\ 0.01 \end{array}$	<0.10
Р3	Pz <sub>up</sub>	$\begin{array}{c} 8.04 \pm \\ 0.02 \end{array}$	$661\pm7$	$\textbf{5.6} \pm \textbf{0.2}$	$140\pm5$	<1.00	$\begin{array}{c} \textbf{7.90} \pm \\ \textbf{0.13} \end{array}$	$0.02\pm0.00$	$\textbf{7.28} \pm \textbf{0.07}$	$0.19 \pm 0.00^{*}$	$\begin{array}{c} \textbf{0.05} \pm \\ \textbf{0.01} \end{array}$	<0.10
	Pzdown	$7.51 \pm 0.02$	$676\pm9$	$\textbf{5.8} \pm \textbf{0.1}$	$155\pm4$	<1.00	9.47 ± 0.20	$0.06\pm0.04$	$\textbf{8.51} \pm \textbf{0.10}$	$0.12\pm0.03$	$\begin{array}{c} 0.10 \ \pm \\ 0.06 \end{array}$	<0.10

\*All measurements are below LOD (0.38 mg/L) hence standard error is 0.00.

#### Table A.4

Weekly applied irrigation volume (WWv (mm)), measured volumetric water contents ( $\theta_i$ ) during the irrigation events (before irrigation ( $\theta_0$ ), at saturation ( $\theta_s$ ) and at field capacity ( $\theta_{FC}$ )) and calculated times for  $\theta_i$  maintenance: drainage time (DT) during which the volumetric water content is above the field capacity and saturation time (ST) during which the volumetric water content is above the  $\theta_s$ .

	WWv (mm)	depth	$\theta_0 (m^3/m^3)$	$\theta_S (m^3/m^3)$	$\theta_{FC} (m^3/m^3)$	ST (h) ( $t_{VWC} >  heta_{S}$ )	DT (h) ( $t_{VWC} >  heta_{FC}$ )
P1	$506.1\pm46.0$	15 cm	$0.305\pm0.001$	$0.331\pm0.001$	$0.322\pm0.001$	$\textbf{3.5}\pm\textbf{1.9}$	$18.3\pm2.6$
		45 cm	$0.288\pm0.003$	$0.335\pm0.001$	$0.319\pm0.002$	$2.0\pm0.6$	$13.3\pm2.0$
P2	$321.7\pm44.5$	15 cm	$0.275\pm0.007$	$0.369 \pm 0.002$	$0.320\pm0.001$	$1.4\pm0.3$	$14.8\pm2.3$
		45 cm	$0.268\pm0.006$	$0.328\pm0.001$	$0.312\pm0.002$	$3.1\pm0.8$	$15.2\pm1.0$
P3	$80.7 \pm 9.4$	15 cm	$0.313\pm0.003$	$0.344 \pm 0.001$	$0.329 \pm 0.002$	$9.3\pm1.7$	$35.1\pm2.6$
		45 cm	$0.295\pm0.002$	$0.315\pm0.001$	$0.309 \pm 0.001$	$\textbf{9.8} \pm \textbf{1.6}$	$30.5\pm3.6$



**Fig. A.1.** Soil moisture contents measured during irrigation events. (A) Weekly volumetric water content measurements for furrow (left) and drip (right) irrigation. (B) Volumetric water content before irrigation ( $\theta_0$ ) at saturation ( $\theta_s$ ). Weekly drainage time (DT) and saturation time (ST) at 15 cm (C) and 45 cm (D), with corresponding applied WWv (E).

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