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Combined natural wastewater treatment systems for removal of organic matter and phosphorus from polluted streams

Bilal Tunçsiper

University of Nigde Omer Halisdemir, Faculty of Engineering, Department of Environmental Engineering, Nigde, 5125, Turkey

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ABSTRACT

Natural wastewater treatment (NWT) systems may be a most suitable and economical solution method to ensure a cleaner environment and sustainable production in control of water pollution. Therefore, in this study, a NWT system was designed to reduce existing pollution in over-polluted Kızılca creek (Nigde, Turkey). The combined system consists of a settlement basin (SB), free water surface constructed wetland (FWS-CW) planted with *Phragmites communis*, and overland flow (OF) system planted with *Italian ryegrass*, respectively. The system was installed on the edge of the creek in 2014. Over-polluted creek water was treated by passing through the system, and the treated water was discharged into the creek. Total suspended solid (TSS), biochemical oxygen demand (BOD), chemical oxygen demand (COD), and total phosphorus (TP) parameters were analyzed on samples taken from the inputs and outputs of all stages of the system. During an operating period of about 18 months, it was observed that the pollutant parameters were reduced to a great extent. Despite the high organic loading, it was determined that the treatment system could remove the TSS and the BOD up to 85%, and the TP up to 49%. Average removals were found to be higher in warm weather conditions where vegetation could grow best. Results suggest that the combined NWT systems can be used as a low-cost wastewater treatment alternative to improve the water quality of polluted streams in similar areas.

1. Introduction

A sustainable clean production can be achieved by treating the wastewater at its source by using economical and environmentally friendly natural wastewater treatment (NWT) systems such as Constructed Wetland (CW) and Overland Flow (OF) systems (Feo and Ferrara, 2017; Yang et al., 2017). The CW and OF systems, which include biotic and abiotic reactions in a single reactor, have been commonly used to remove pollutants in urban, industrial, agricultural, and stormwater run-off (Kadlec and Wallace, 2009; Upadhyay et al., 2016; Greenway, 2017).

Prior applications for improving water quality in polluted streams usually included improvement of water quality through wastewater treatment systems established near streams (Tu et al., 2014) or stream rehabilitation (Hunt et al., 1999; Richardson et al., 2011; Stone et al., 2003). Hunt et al. (1999) and Stone et al. (2003), have applied in-stream wetlands in order to restore or remediate the polluted stream due to non-point source pollution (NSP). On the other hand, Richardson et al. (2011), examined water quality in Upper Sandy Creek in North Carolina.

Researchers inferred that the restoration or the rehabilitation application of the polluted streams or the improvement of ecosystems in streams might reduce pollution. Tu et al. (2014), evaluated the effectiveness of the CW systems located on the side of Kaoping river in Kaohsiung (Taiwan) on the treatment of polluted Kaoping. Results showed that the CWs had a grand potential to improve water quality. On the other hand, some researchers (Helfield and Diamond, 1997; Mutiti et al., 2015) have emphasized that the CW systems used for in polluted river water purification had a limited treatment capacity. For example, Helfield and Diamond (1997), have evaluated results obtained from a project proposed for the Don River that discharged at the northwestern shore of Lake Ontario, and they have emphasized that the CW system had a limited removal efficiency (RE) and remained incapable in organic matter and the P removal.

Kızılca (Karasu) creek, which is the main topic of this study, is heavily polluted due to high organic matter and phosphorus-containing septic discharges, untreated discharge waters of treatment plants, agriculture runoff, and illegal discharge waters. The

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E-mail address: tuncsiperb@ohu.edu.tr.

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creek feeds Akkaya reservoir that is one of the most important useable water sources of the Nigde City (Turkey). Furthermore, in this study, a NWT system that is environment-friendly, easily applicable and economical was considered as an alternative method to improve water pollution in polluted streams. The study that proposes a new system where the CW system with filtering materials was used in conjunction with the SB and the OF system contributes to existing knowledge. Consequently, the study will be considered as a reference for decision makers and researchers in further scientific studies as it was first applied as an alternative method in our country to control water pollution in over-polluted creeks or streams.

2. Materials and methods

2.1. Study area

The most important pollutant sources (see features denoted 1, 2, 3 and 4 on Fig. 1) that pollute the dam, the position of the creek and the NWT system are shown in Fig. 1. Important point sources threatening dam are discharge waters of Organized Industrial Zone Wastewater Treatment Facility (OIZWTF), Nigde University Wastewater Treatment Facility (NUWTF), Nigde Municipality Wastewater Treatment Facility (NMWTF), and Kızılca creek. The creek is one of the most important feeding (with an average flow rate of 0.43 $m^3 s^{-1}$) and polluting sources of the dam (Nigde, Turkey). The creek is about 27 km long within the borders of Nigde City, and it flows into the dam.

Effluents of the OIWTF and NUWTF were discharged to the dam from the campus area of the Nigde University. In addition, the creek was also under the influence of surface water draining from agricultural fields, lime quarry-pit, and outflows of the NMWTF, which reached the creek. Because the NMWTF that treats wastewater of about 120000 populations was not operated very well, a large amount of pollutant reached the dam through the creek.

The NWT system was located on the edge of the creek, and in a region at the coordinates- 37°56′23.20″N and 34°39′13.05″E of Nigde City, Turkey. The distance between the dam and the NWT system was about 4.5 km along the creek length. The NWT system was established in August 2014 and operated for a period of about 18 months.

2.2. Description of the combined NWT system

Since the NMWTF was not operated very well, its discharge waters further increased the pollution in the creek. Therefore, the combined NWT system was built approximately 600 m downstream the creek from the pollution point where the exit waters of the NMWTF were discharged.

The NWT system, which consists of four stages, is planned in a manner appropriate to the creek' native structure (Fig. 2). The first stage comprises a feeding basin (FB), the second stage comprises Settlement Basin (SB), the third stage comprises Free Water Surface Constructed Wetland (FWS-CW) system, and the fourth stage comprises OF system.

The FB system was designed to feed the NWT system, and to prevent clogging of the FWS-CW system. The SB system was designed to remove organic matters by sedimentation and thus increase the FWS-CW and the NWT system's treatment efficiency. The FWS-CW system was designed to remove the TSS, particulate and especially dissolved organic matter, and inorganic phosphorus. Dissolved organic matters, which cannot be removed in the SB system, is removed by mostly aerobic oxidation, partly bacteria, and plant uptake. Organic matters and inorganic phosphorus is removed by bacteria and plant uptake, absorption, and by adsorption onto depositing sediments in the FWS-CW system. The FWS-CW system was designed S-shaped in a manner to represent the convoluted structure of the creek.

In order to further improve the RE of organic matters and phosphorus, in April 2015, twist places of the system were equipped with a filtration layer that serves as a biofilter. The grain diameter of gravel material used for the filtration layer ranges 0.8–6.3 cm. The total volume of gravel used in the system is approximately 0.6 m^3. System was planted with the young shoots of *Phragmites communis* (macrophytes) growing in the creek edge; and (iv) the OF system, which was consisted of washed sand in a depth of about 5 cm, was designed to be able to provide an extra organic matter and phosphorus removal in outflows of the FWS-CW system. It was planted with *Italian ryegrass*.

The distance between the FB and SB is 1.55 m. The total water volume of the system is approximately 14.5 m^3. In the FWS-CW system, the percent volume occupied by the filtration material and the plants are approximately 1% and 19%, respectively. The

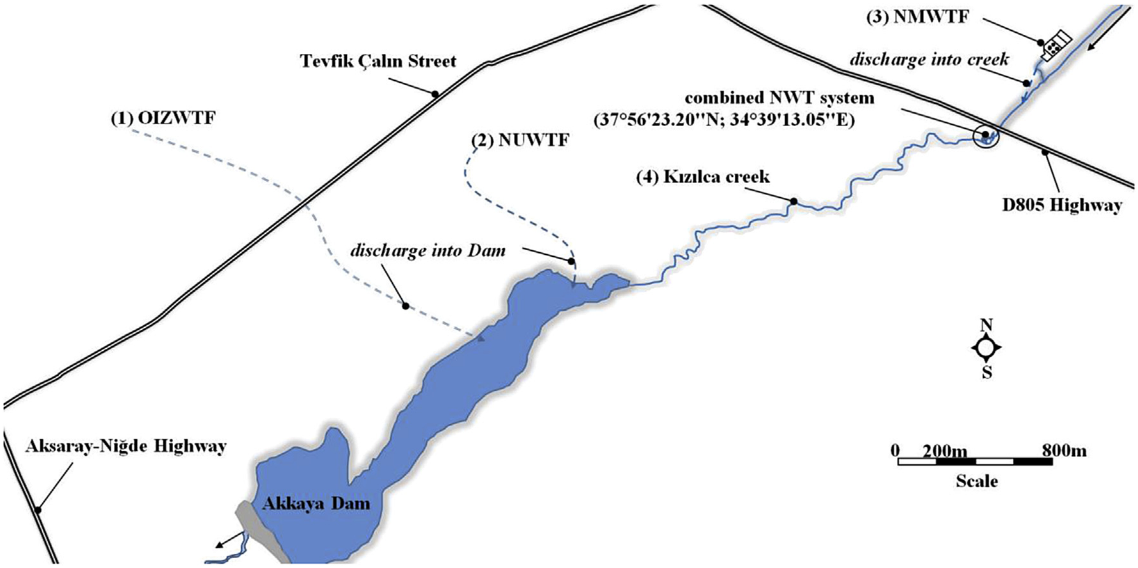

Fig. 1. Akkaya dam and location map of pollutant sources it pollutes.

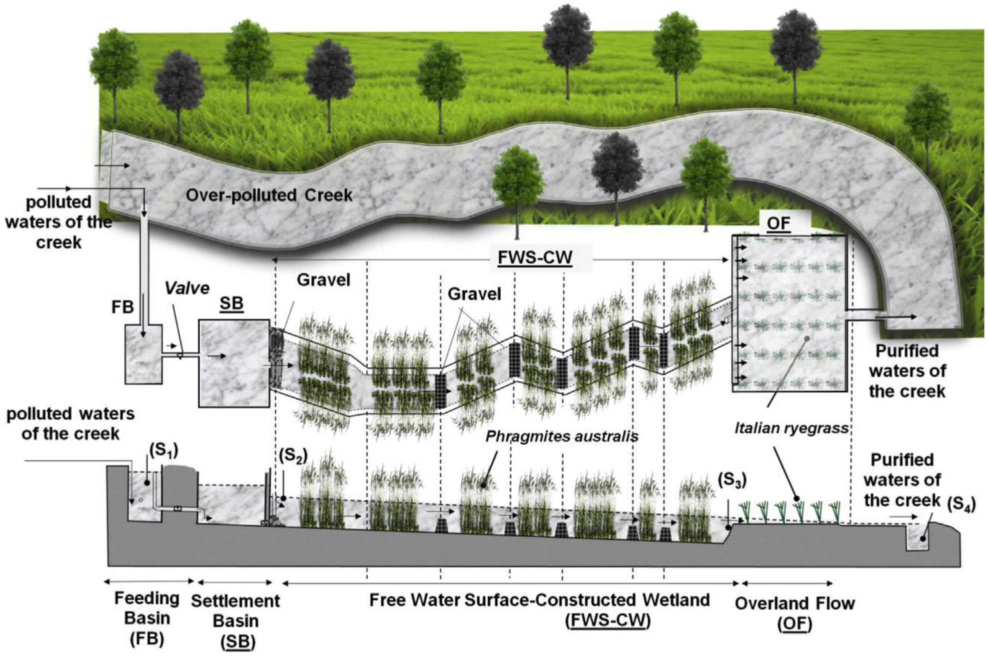

Fig. 2. Schematic diagram of the NWT (S_1, S_2, S_3, S_4 are sampling locations) system.

porosity of the filtration material used in the system is the ratio of the volume of voids in the filtration material to its total volume, and it is about 0.53 (dimensionless). The total length of the NWT system is approximately 57 m. A detailed account of the NWT design is summarized in our previous study concerning nitrogen removal from polluted streams by using hybrid NWT systems (Tunçsiper, 2018).

2.3. Sampling and physicochemical analysis

Water samples were collected and analyzed semimonthly or monthly from 2014 to 2016 to evaluate the effectiveness of the NWT system on pollutant removal, and thus the improvement of water quality of the creek. In this study, water samples were collected from four sampling locations including S_1 (influent of SB system), S_2 (effluent of the SB system or influent of the FWS-CW system), S_3 (effluent of the FWS-CW system or influent of the OF system), and S_4 (effluent of the OF system) (Fig. 2). Average number (n) of samples were 13 for each of the sampling locations.

Turkish Water Pollution Control Regulation (TWPCR) (TWPCR, 2004) and International Organization for Standardization 5667–3 (ISO, 2013) were taken as a basis in sampling, and in preservation and analysis of the samples. 2-hour composite samples were prepared for analysis. Sampling points and numbers were determined in a manner to characterize water quality in the sampling area and the variation of water quality within treatment systems.

Temperature, Dissolved Oxygen (DO), Electrical Conductivity (EC) and pH were determined in situ with a WTW inolab-IDS multi 9430 parameter probe. The measurement was obtained by immersing the probes of the testing machine inside the samples. Biochemical oxygen demand (BOD) was analyzed using a close respirometric unit WTW OxiTop IS 6. OxiTop was based on pressure measurement in a closed system. The CO_2 formed in OxiTop was absorbed by NaOH to form a vacuum and the resulting vacuum was measured as mg BOD/L.

The amount of available oxygen for BOD is adjusted according to the sample volume used. Total suspended solid (TSS), total organic carbon (TOC), chemical oxygen demand (COD), and total phosphorus (TP) were performed following Standard Methods (APHA, 1998).

The TSS was determined by using gravimetric technique. The sample water was filtered through a filter paper of known weight. The paper was dried out and weighed. Then the filter paper was heated at 105 °C and weighed again. The TOC was determined by high-temperature combustion method. After the sample was acidified, the CO_2 generated is determined, and then the TOC was calculated by subtracting the total inorganic carbon from the total carbon. The COD was determined by the titrimetric method. After standard potassium dichromate digestion solution was prepared, digestion solution and reagent water (H_2SO_4) was added to sample and then it was titrated with standard ferrous ammonium sulfate titrant by using ferroin indicator. Total phosphorous (TP) was determined by the vanadomolybdophosphoric acid colorimetric method (SM-4500-P C).

After pH adjustment and color removal, the vanadate-molybdate reagent was added to the samples, and samples were diluted to the mark with distilled water. Absorbances of samples were measured versus a blank at a wavelength of 400–490 nm.

2.4. Statistical analysis

Statistical analysis was carried out using one-way ANOVA, paired-samples *t*-test and univariate-general linear model on the SPSS analytical software (IBM SPSS Statistics Software, 2016) to assess the effect of the TSS, the BOD, the COD, and the TP loading rates and different weather conditions (cold and warm weather conditions) on treatment efficiencies of the systems, and to check whether or not the difference between the inlet and outlet concentrations was statistically significant. Pearson's correlation was used to evaluate relationships between variables based on p-values at the 0.05 level.

2.5. Removal rate constants (RRCs)

There are many models which express the temporal change of organic matter and nutrient RRC depending on operating parameters in the CWs and OF systems. Many researchers (Birgand et al., 2007; Kadlec and Knight, 1996; Kadlec et al., 2000) concluded that the first-order model was a better choice because the hydraulic flow conditions in the CWs and OF systems were very similar to plug-flow conditions.

In this study, the RRCs of the organisms at 20 °C (k_{20}) were calculated by an equation (1)which showed a first-order reaction kinetic given based on the relationship between hydraulic residence times (HRTs) and influent and effluent concentrations (Kadlec and Knight, 1996; Kadlec et al., 2000).

$$\frac{C_e - C^*}{C_i - C^*} = \exp[-(KT_v)(t)] = \exp\left[-\left(\frac{KT_A}{h}\right)\left(\frac{1}{\varepsilon}\right)\right] \quad (1)$$

Where; C_i, C_e, and C^* are influent, effluent, and background concentrations (mg L^{-1}), respectively. “h” is water depth (m). KT_A is a real rate constant (m d^{-1}) at temperature T °C, and $K_{A(20)}$ is removal rate constant (m d^{-1}) at 20 °C. KT_V is volumetric rate constant (d^{-1}) at temperature T °C, and $K_{V(20)}$ is rate constant (d^{-1}) at 20 °C. $KT_A = k_{A(20)}\theta^{T-20}$, and $KT_V = k_{V(20)}\theta^{T-20}$. “$t$” is hydraulic residence time (HRT) (d). “ε” is porosity. “θ” is temperature correction factor. “T” is water temperature (°C)≈0. In order to compare the theoretical removals in all stages of the pilot system to the RRCs which are the indicators of the RRCs by microorganisms in the ($\frac{C_o - C_e}{C_o} x100$) system.

3. Results and discussion

3.1. Variation in the pH, the DO and the EC values of the current stages of the NWT system

Flow rate, water temperature, the pH, the DO and the EC values of the current stages in the NWT system are shown in Table 1. Non-point sources entering the creek through run-off and rainwater or point sources such as effluents of treatment plants influenced the pH, the DO, and the EC values in the creek and NWT system. While the DO decreased, the EC values increased with the increase of organic matter (TSS, BOD, and COD) load in the influent water.

The EC is the sum of the salt in water or soluble substances. The EC values in the system exceeded 1000 μS cm^{-1} at high organic loading. However, The EC values in rainy days decreased up to 945 μS cm^{-1} in the creek and 723 μS cm^{-1} in effluents of the system. In the literature, it was emphasized that the EC values in rainy days might decrease possibly associated with dilution effect of rainwater on pollutants or settlement of the dissolved organic matters (Crites et al., 2004; Kadlec and Wallace, 2009).

The FWS-CW system provides additional oxygen sources for decomposition of organic matters because of atmospheric reaeration and submerged vegetation (Crites et al., 2004; Kadlec and Wallace, 2009; USEPA, 2000). For these reasons, the DO concentrations in the effluent of the FWS-CW system were found to be higher than influent (Table 1). The possible reasons of the DO increase in the CW and OF systems may be due to the plants and the occurrence of oxygen transfer from the water surface (Ariyanti et al., 2011; Kadlec and Wallace, 2009; Tu et al., 2014).

The pH in the FWS-CW system increased from 7.29 in the influent to 7.60 in the effluent (Table 1). The pH values in the CW systems may increase probably due to photosynthesis process or stored mud in the bottom of system exposure to anaerobic conditions (Ariyanti et al., 2011; Tu et al., 2014).

3.2. The water quality of the creek

Whereas concentrations in the creek about 250 m upstream of the NWT system were 532 mg L^{-1} for the TSS, 422 mg L^{-1} for the BOD, 44.4 mg L^{-1} for the COD, and 5.5 mg L^{-1} for the TP, these concentrations about 300 m downstream of the NWT system were found to be 336 mg L^{-1} for the TSS, 343 mg L^{-1} for the BOD, 39.6 mg L^{-1} for the COD, and 5.1 mg L^{-1} for the TP.

Especially organic matter concentrations in the downstream slightly decreased probably due to the dilution of organic pollutants along the creek’ length. Because the percentage of the flow rate of the effluents of the NWT system to the total flow rate of the creek was quite low, the impact of the effluents of the NWT system on the water quality of the creek will be negligible. For these reasons, in this pilot-scale study, it is aimed to purify only a certain part of the creek rather than the entire flow.

3.3. The RRC-removal efficiency (RE) relationship

The second-order polynomial regression was obtained from relationships between log K_T and T-20. The RRCs (k_{20}) were obtained from the second-order polynomial regressions. The values for k_{20} (day^{-1}) and the average RE (%) were 0.26 and 63 for the TSS, 0.32 and 64 for the BOD, 0.29 and 56 for the COD, and 0.13 and 30 for the TP in the FWS-CW system respectively, although they were 0.18 and 31, 0.20 and 31, 0.19 and 29, and 0.08 and 16 in the OF system respectively, and lower. These results indicated that the RRCs were related to the designs and the REs of the systems.

The k_{20} value (0.26 day^{-1}) for the TSS in the FWS-CW system was closer to the values reported (0.28–0.68 d^{-1}) by Kadlec and Knight (1996). The BOD of 0.32 day^{-1} was lower than that (0.40 d^{-1}) reported by Kadlec and Knight (1996). The k_{20} values for the BOD obtained by Jing et al. (2002) varied between 0.38 and 0.55 day^{-1}. Reed et al. (1988) suggested the k_{20} value of the BOD as 0.49 d^{-1} for industrial wastewaters with high COD. The k_{20} value of The COD was slightly lower than the value 0.49 d^{-1} obtained by Sawaittayothin and Polspart (2006).

In addition, many researchers noted that the k_{20} value for the TSS and TP was independent of temperature. Although the k_{20} value for the TP rate was found to be lower than the value 0.28 d^{-1} recorded by Wittgren and Maehlum (1997), it was very close to the value 0.14 d^{-1} recorded by Kadlec and Knight (1996). It was estimated that these differences in the rate constants might result from especially different designs, different wastewater characteristics, different loading rates, and environmental conditions.

Table 1
Water quality data and flow rate of the NWT system.

Parameters (the average values for the 18 months of system operation)	SB influent	SB effluent or FWS-CW influent	FWS-CW effluent or OF influent	OF effluent
Flow rate (m^3 d^{-1})	1707 ± 1369	1680 ± 1342	1565 ± 1268	1534 ± 1252
Water temperature (°C)	11.36 ± 3.67	11.12 ± 3.70	10.11 ± 3.62	9.59 ± 3.41
pH	7.32 ± 0.26	7.29 ± 0.29	7.60 ± 0.34	6.91 ± 0.38
DO (mg L^{-1})	0.34 ± 0.14	0.31 ± 0.16	1.41 ± 0.25	0.41 ± 0.26
EC (μS cm^{-1})	1091 ± 81	1036 ± 72	950 ± 87	898 ± 120
Organic matter load (g m^{-2} d^{-1})*	571.6 ± 332.2	157,1 ± 92,1	3.08 ± 2.14	0.84 ± 0.57

Note: ±means standard deviation, and * includes TSS, BOD, and COD.

3.4. *Effect of the seasonal variations*

At the beginning of the system's operation period (between October and November), the growth of macrophytes in the FWS-CW and OF system was adversely affected by cold weather conditions. In addition, on the dates specified, the TSS, the BOD, and the COD loading increased also considerably. In cold weather conditions (January—December), the RE of the FWS-CW system decreased probably because both the macrophytes gradually died completely and bacterial activity decreased. When ice covered open water, the transfer of oxygen from the atmosphere was reduced, decreasing oxygen and temperature-dependent carbon oxidation (Kadlec and Wallace, 2009).

On the other hand, in warm weather conditions (from April to August), the REs of the FWS-CW and OF system for the TSS, the BOD, and the COD gradually increased. The results indicated that the macrophytes and rising temperature rise helped greatly in removing solids from the water. The macrophytes provide surface area for colonization by bacteria that contribute to the processing of carbon and other wastewater constituents (Kent, 2001).

The TSSs are removed from the water column of the FWS-CW system by flocculation, sedimentation, filtration, adsorption, and plant and bacteria uptake and degradation. In addition, filtration between plant stems and sedimentation in these systems is highly effective because of low flow velocity, and thus a longer retention time (Hammer et al., 1993; Kadlec and Knight, 1996; Karathanasis et al., 2003). The removal of settleable organics (the TSS, the BOD, and the COD) is very rapid and high (>90%) in the FWS-CW systems, possibly associated with quiescent conditions and deposition. However, the OF systems have a lower purification volume and filtration area for the TSS and BOD removal because that the TSS and BOD removal essentially occurs in regions close to the only soil surface and in plant matrix (Crites et al., 2004; USEPA, 2006).

In this study, although variations between average TSS, BOD, and COD influent and effluent concentrations increased in the FWS-CW system towards the end of the study period depending on age, these variations decreased in the OF system. These results showed that the FWS-CW system providing the best development and growth of plants and bacteria had a higher organic matter removal compared to the OF system, probably due to a higher microbial degradation and particulate settling and denser vegetation (Kadlec and Wallace, 2009). The average percent REs, the correlation coefficients (R^2) and the p-values of the current stages of the NWT system in cold and warm weather conditions are shown in Table 2.

As shown in Table 2, the REs of the FWS-CW system in warm weather conditions were found to be about higher 16% for the TSS, 22% for the BOD, and 12% for the COD compared to cold weather conditions, although they were at about the same levels for the SB and OF system. On the other hand, the RE of the TP was found to be about 3% higher for warm weather conditions in all systems. Average RE of the FWS-CW system during warm weather conditions was higher than other systems, probably due to a higher treatment volume, and thus higher sedimentation and decomposition (Table 2).

The p-values less than 0.05 and positive correlations indicated that seasonal differences were significant on the REs of the organic matter (except for the TSS in the SB system) and the TP. The p-values higher than 0.05 and negative correlations indicated that seasonal differences were not significant on the RE of the TSS in the SB system.

In the literature, the reports on the influence of weather conditions on the removal of pollutants in the CW and OF system were not unanimous (Crites et al., 2004; Kadlec and Wallace, 2009). For example, Kadlec et al. (2003), reported that the highest organic matter (the TSS, the BOD, and the COD) and the TP removals of the CWs were during autumn and summer with a large decrease during winter and spring. Smith (1982), reported that the average removals of the OF systems were higher during winter weather conditions (Crites et al., 2004).

Vymazal (2001), reported that there was a very low difference in organic matter removals between summer and winter in the CWs. Brix (2007), reported that there was a very low the effect of seasonal variations on the REs of the CWs. Jenssen and Maehlum (2003), found that there was no considerable difference in the RE of the BOD and TSS between cold and warm periods in nine CWs in Norway.

3.5. *Removal rate (RR)-organic loading rate (OLR) and phosphorus loading rate (PLR) relationship*

Average RRs and percent REs in the individual beds (first the SB, second the FWS-CW, and third the OF) operated in series based on average influent and effluent concentrations, the HRTs, hydraulic loading rates (HLRs), the OLRs, and the PLRs are shown in Table 3. The relationships between loadings and their RRs were found to be linear but exponential for only the TP in the OF system.

Table 2
Average percent REs in cold and warm weather conditions.

			SB	FWS-CW	OF
TSS	In cold weather conditions	Average removal (%)	45	54	31
	In warm weather conditions	Average removal (%)	47	70	32
		R^2	−0.14	0.52	0.81
		p-value	0.233	0.001	0.014
BOD	In cold weather conditions	Average removal (%)	37	54	29
	In warm weather conditions	Average removal (%)	40	76	30
		R^2	0.47	0.57	0.51
		p-value	0.019	0.002	0.003
COD	In cold weather conditions	Average removal (%)	40	50	28
	In warm weather conditions	Average removal (%)	40	62	29
		R^2	0.83	0.57	0.57
		p-value	0.007	0.001	0.004
TP	In cold weather conditions	Average removal (%)	13	28	13
	In warm weather conditions	Average removal (%)	16	34	18
		R^2	−0.084	0.89	0.87
		p-value	0.78	0.000001	0.005

Table 3
Average influent and effluent concentrations, the HRT, the HLR, the LR, the RR, and the RE of the NWT system.

		Influent	Effluent	HRT	HLR	OLR	PLR	RR	RE
		mg L^{-1}		day	$m^3\ m^{-2}\ d^{-1}$	g $m^{-2}\ d^{-1}$			%
TSS	SB	479 ± 233	255 ± 132	3.64	1.52	823		386	46
	FWS-CW	255 ± 132	95 ± 100	10.7	4.80	199		85	55
	OF	95 ± 100	66 ± 71	1.23	0.69	41		11	30
BOD	SB	375 ± 221	234 ± 141	3.64	1.52	664		222	37
	FWS-CW	234 ± 141	85 ± 99	10.7	4.80	200		100	58
	OF	85 ± 99	59 ± 72	1.23	0.69	37		10	29
COD	SB	569 ± 393	344 ± 229	3.64	1.52	1098		405	40
	FWS-CW	344 ± 229	150 ± 160	10.7	4.80	315		136	58
	OF	150 ± 160	107 ± 122	1.23	0.69	67		18	28
TP	SB	6.4 ± 2.9	5.4 ± 2.5	3.64	1.52		4.97	1.17	13
	FWS-CW	5.4 ± 2.5	3.8 ± 2.0	10.7	4.80		3.80	0.70	21
	OF	3.8 ± 2.0	3.2 ± 1.9	1.23	0.69		1.20	0.07	10

Note± means standard deviation.

One-way ANOVA test results showed that the OLRs (the TSS, the BOD, and the COD) and their RRs had significant correlations (all $p < 0.05$). The correlation coefficients ($R^2 > 0.84$) between loadings (g m^{-2} d^{-1}) and their RRs (g m^{-2} d^{-1}) showed that the OLRs and PLRs had a positive impact on their RRs. While RRs increase together with the increase of the PLRs in the SB and FWS-CW system, the RRs of the TP in the OF system tended to decrease and reached a stable constant value. Functional decreases for the RRs of the TP in the OF system depending on the PLRs observed when the PLRs exceed about 30 g m^{-2} d^{-1}. These results showed that the RRs of the TP in the OF system were adversely affected by high PLRs during the operation period.

In this study, while the FWS-CW system was operated in much higher BOD loading (average 200 g m^{-2} d^{-1}) than the values (average 10 g m^{-2} d^{-1}) reported in the literature, the OF system was operated in the range of values (3.5–11 g m^{-2} d^{-1}) in the literature (Crites et al., 2004; USEPA, 2006). The loadings recommended for achieving target TSS and BOD effluent concentrations between 20 and 40 mg L^{-1} in the FWS-CWs were given between 3 and 7 g m^{-2} d^{-1} (Wallace and Knight, 2006; USEPA, 2000). Wallace and Knight (2006) pointed out that the P removal of the FWS-CWs were not very effective in higher inflow PLRs than 0.1 g m^{-2} d^{-1}. Although the FWS-CW and OF system fed at substantially higher loadings than the values recorded in the literature, the average RE was quite satisfactory.

3.6. The RE-the HLR relationship

There were typically exponential decreases in the RE with increasing the HLR in the FWS-CW and OF system. The REs reached a stable (constant) value by increasing the HLRs. This may be due to the fact that the increases in the HLRs increase the amount of pollutants passing through the system and the pollutant accessibility to microorganisms. The REs were at approximately the same level during all operation period in the SB system. The FWS-CW system has provided higher REs despite higher LRs than others. This could be explained by the relatively larger treatment volume, larger plant and bacterial activity, and lower HLR (Harrington and Ryder, 2002).

Test results showed there was a statistically significant relationship between the HLR and the RE ($p < 0.05$). Results were generally consistent with the literature. A vast majority of researchers concluded that the REs of the TSS, the BOD, and the COD decreased with the HLRs (Calheiros et al., 2009; Crites et al., 2004; Kadlec and Wallace, 2009). The FWS-CW system exhibited a plateau, or background TSS removal of 20%, the BOD and COD removal of 40%, and the TP removal of 15% at the HLRs between 1.0 and 1.7 m^3 m^{-2} d^{-1}. Additionally, the OF system exhibited a background TSS, BOD, and COD removal of 25%, and TP removal of 4% at the HLRs between 0.3 and 0.6 m^3 m^{-2} d^{-1}.

The HLRs reported for large-scale FWS-CW systems in the literature vary greatly and they are much lower than the HLR applied in this study. The HLRs were commonly given in the range of 1–100 m yr^{-1} (≈0.003–0.3 m^3 m^{-2} d^{-1}) for the FWS-CWs (Crites and Tchobanoglous, 1998), and to be approximately 10 m yr^{-1} (≈0.03 m^3 m^{-2} d^{-1}) for the OF systems (Crites et al., 2004; USEPA, 2006).

In this study, despite the NWT system was operated in extremely high hydraulic loads (0.1–1.7 m^3 m^{-2} d^{-1} or 10–170 cm d^{-1}), and a much higher average influent TSS concentration of 563 mg L^{-1} and BOD concentration of 471 mg L^{-1}, the average removals (82%) of organic matter were quite satisfactory. In previous studies, the average REs of the CWs were recorded between 28% and 94% for the less polluted streams (Kadlec and Wallace, 2009; Wen et al., 2007).

Wen et al. (2007), tested the TSS, the BOD, the COD, and the TP removals on an OF system consisting of eight channels to treat the less polluted creek water. At an average HLR of 0.278 m^3 m^{-2} d^{-1} and at low influent TSS concentration of 52 mg L^{-1}, the BOD concentration of 12 mg L^{-1}, and the COD concentration of 73 mg L^{-1}, and the TP concentration of 2.4 mg L^{-1}, the OF system removed 78% TSS, 42% BOD, 48% COD, and 33% TP.

In this study, although the HLR applied on the OF system was higher (average 1.52 m^3 m^{-2} d^{-1}), the OF system removed 31% TSS, 29% BOD and COD, and 16% TP at a much higher average influent TSS concentration of 95 mg L^{-1}, the BOD concentration of 85 mg L^{-1}, the COD concentration of 150 mg L^{-1}, and the TP concentration of 3.8 mg L^{-1}.

3.7. The RE-the HRT relationship

Apparent relationships between the average effluent concentrations and the HRTs are shown in Fig. 3. Test results showed the HRTs that had a significant effect on the average effluent concentrations and the REs ($p < 0.05$).

As shown in Fig. 3, it is seen that the average effluent concentrations for the FWS-CW and OF system initially increase along with the increase of the HRTs, but then they decline. Cui et al. (2015), found similar results for P removal in CWs. On the other hand, Kadlec and Wallace (2009), reported a linear decrease in effluent TSS, BOD, and COD concentrations with increasing loading in the FWS-CWs.

Optimum HRT appears to be about 1.0 days for the OF system, 2.0 day for the SB system, and 4.0 day for the FWS-CW system. The

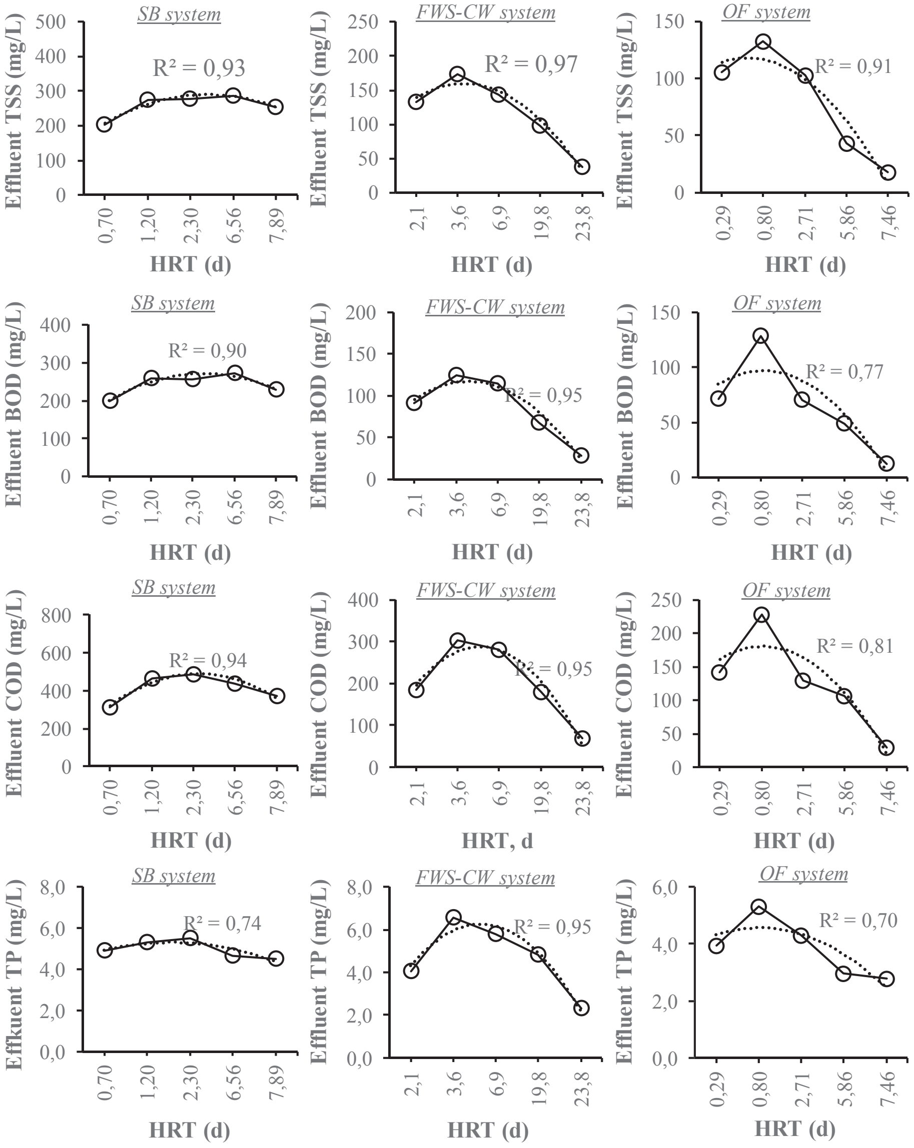

Fig. 3. The average TSS, BOD, COD, and TP effluent concentrations as a function of the HRT.

FWS-CW system, which has the highest HRT value, has the highest RE. These results show that the HRT has a significant effect on the REs of the systems. Bacteria need a certain period of time (optimum HRT) to be able to oxidize the whole of the organic matters entering the system. If this time period is not sufficient for bacteria to decompose all organic matters, a certain portion of the organic matters in the system can leave the system without being oxidized. This can significantly reduce the RE of the system.

3.8. The effect of filtration material on the RE

In order to determine the effect of filtration on the REs, twist places of the FWS-CW system, which form the basis of the treatment system, were equipped with filtration material (gravel) in periods between April and August towards the end of operation period. The REs in the operating periods where the HLR was constant was shown in Table 4.

Test results revealed that the average REs of the FWS-CW system after and before the use of the filtration material were statistically different ($p < 0.05$). At the same HLR, average REs of the FWS-CW system after the use of the filtration material were found to be higher than those before the use of filtration material. As shown in Table 4, when the NWT system was operated in approximately the same HLRs, the REs of the FWS-CW system increased approximately 20% for organic matters (the TSS, the BOD, and the COD), and 4% for the TP on average.

Table 4
Average REs of the treatment systems before and after the use of filtration material.

		Average percent REs in the same HLR	
		FWS-CW system	NWT system
Before the use of filtration material	TSS	70	89
	BOD	76	91
	COD	52	79
	TP	23	44
After the use of filtration material	TSS	88	96
	BOD	90	97
	COD	86	95
	TP	27	48

Similarly, depending on the success of the FWS-CW system, the REs of the NWT system increased approximately 10% for organic matters, and 2% for the TP on average. These results indicated that the use of the filtration materials might improve especially organic matter REs in the NWT system (Lu et al., 2016; Saeed et al., 2018).

3.9. The RE of the NWT system

The FWS-CW system has a higher capacity than others in the TSS, the BOD, and the COD removal (Table 3). As shown in Table 3, the removal of organic matter and phosphorus in the planted FWS-CW system was better than the unplanted SB system. This indicated that the plants made an important contribution to organic matter removal, probably due to the plant uptake and adsorption, and the that the roots of plants and the filter bed (gravel) provided a biologically active surface area for the biochemical transformation of contaminants and physical processes such as filtering and entrapment of particulates (Kyambadde et al., 2004; Li et al., 2009; Riggio et al., 2018). The attached bacteria on the filter bed and the roots of the plants remove a large part of dissolved organic matters by biodegradation.

During the study period, the average BOD/COD ratio, which indicates the biodegradability of organic substances, was about 0.68 at the inflow of the FWS-CW system, while it was 0.56 in the outflow. This result indicated that biodegradable organic matters was decreased at the system's outflow.

The average TSS and BOD removals were approximately 63% in the FWS-CW system and 29% in the OF system. Compared to the TSS and BOD removals of 85%–95% recorded for the FWS-CW and the OF systems in the literature (Crites et al., 2006; Kadlec and Wallace, 2009; Vymazal, 2010), the possible reason for these lower removals might be due to higher loadings. Although the abilities of each system to remove the TSS and BOD within the NWT system were low, generally, in spite of the higher HLRs, the success of the NWT system in the TSS and BOD removal was high with an average 83% (Table 3).

Bacteria in the CW and the OF system can degrade organic matter within a certain period of time (or the HRT). Due to high HLR, when highly organic matter enters the system, bacteria in the system will not degrade the whole of these matters as the HRT will be shortened. Therefore, the RE of the system will probably decrease (Crites et al., 2006; Kadlec and Wallace, 2009).

In addition, results obtained from Table 3 showed that the use of an SB system as pretreatment might be very useful for the production of lower organic matter and phosphorus effluents. The reasons of low RE for the TP in all the stages of the NWT system might be due to: (1) especially the deficiency of filtration material (such as gravel) that would immobilize phosphorus in treatment media and sediments; (2) anoxic conditions in the bottom bed sediments.

In the literature, it has been stated that the TP removal was limited to about 30–50% (Crites et al., 2006; Kadlec and Wallace, 2009; USEPA, 2006), probably due to a very low contact surface between the soil and applied wastewater, and a lower retention time for sorption in the OF systems, the lack of filtration material in the FWS-CW systems.

In addition, the P can be removed only by physical or gravitational precipitation in the SBs (Tchobanoglous et al., 2004). For those reasons, mostly sedimentation and partly the plant and bacterial uptake is the most important process that removes phosphorus in the NWT system. In this study, although the FWS-CW and OF system could remove the TP up to 30% and 16% respectively (Table 3).

As shown in Table 3, despite the high HLR of approximately 2.3 $m^3\ m^{-2}\ d^{-1}$ (2300 $L\ m^{-2}\ d^{-1}$), approximately 84% of the organic matters (the TSS, the BOD, and the COD) and 49% of the TP were removed in the NWT system.

The NWT system was able to produce organic matter concentrations below the discharge criteria (200 $mg\ L^{-1}$ for the TSS, 75 $mg\ L^{-1}$ for the BOD, 150 $mg\ L^{-1}$ for the COD, and 2 $mg\ L^{-1}$ for the TP) to the receiving waters (TWPCR, 2004). Consequently, the research results indicated that the NWT systems might significantly improve the water quality in polluted streams. So, the study may be used as an alternative method to control water pollution in polluted streams.

4. Conclusions

In this study, a combined NWT system consisting of an unplanted SB followed by planted FWS-CW and OF beds respectively was designed for the polishing of organic matter and phosphorus effluents from polluted streams. In general, the system provided efficient results for controlling water pollution in an over-polluted creek.

The planted systems provided a higher organic matter and phosphorus removal compared to the SB system without vegetation. The FWS-CW system in the second stage provided a higher organic matter removal than the OF system in the last stage. In addition to this, the use of the SB system as a pre-treatment stage has been quite useful to decrease the effect of organic load on the FWS-CW and OF system through sedimentation and filtration.

The removal efficiency of the FWS-CW system in warm weather conditions was found to be about higher 26% for the TSS and BOD and 12% for the COD compared to cold weather conditions, although they were at about the same levels for the SB and OF system. On the other hand, the TP removal efficiency was found to be about 3% higher for warm weather conditions in all systems.

The average removal efficiency of the FWS-CW system during warm weather conditions was higher than other systems, probably due to a higher treatment volume, and thus higher sedimentation and decomposition.

There was a statistically significant relationship between the HLR and the RE. The REs of the TSS, the BOD, and the COD decreased with the HLRs. The HRTs had a significant effect on the average effluent concentrations and REs. The average effluent concentrations initially increased along with the increase of the HRTs, but then they declined.

The results indicated that the RRCs were related to the designs and the REs of the systems. Although the organic matter and phosphorus RRCs in the planted systems were larger than the unplanted SB system, the RRCs in the planted FWS-CW system with higher RE were higher to compared the planted OF system.

The use of filtration material in the FWS-CW system positively affected the RE of the NWT system. After the use of the filtration material in the FWS-CW system, the removal efficiency of the combined NWT system increased approximately 10% for organic matters, and 2% for the TP on average.

The NWT system decreased the TSS from 563 $mg\ L^{-1}$ to 96 $mg\ L^{-1}$, the BOD from 471 $mg\ L^{-1}$ to 88 $mg\ L^{-1}$, the COD from 777 $mg\ L^{-1}$ to 163 $mg\ L^{-1}$, and the TP from 6.2 $mg\ L^{-1}$ to 3.8 $mg\ L^{-1}$. This indicated that the organic matter and phosphorus removal processes (biological, physical, and chemical) occurred simultaneously in the NWT systems, despite the high HLR of average 2.3 $m^3\ m^{-2}\ d^{-1}$.

Finally, research results indicated that the NWT system might be a very successful treatment application for rehabilitation of the stream ecosystem and a most suitable and economical solution method for control of water pollution in developing countries.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2019.04.211.

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