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## Integrated constructed wetlands treating industrial wastewater from seed production

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#### **Abstract**

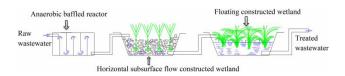
The performance of an integrated wastewater treatment system composed of horizontal subsurface flow constructed wetland (HSSFCW), floating constructed wetland (FCW), and anaerobic baffled reactor (ABR) was studied for pollutant removal from seed production wastewater. *Cyperus alternifolius (Umbrella Papyrus) plants* were used in the HSSFCW, and *Vetiveria zizanioides* (Vetiver grass) in the FCW. The ABR was fed with 25 m³/d wastewater from its equalization tank. The average raw wastewater organic loading rate was 0.208 kg-COD/d. Grab wastewater samples were collected twice weekly for three months from each unit's inlet and outlet. The system's performance in removing biochemical oxygen demand (BOD<sub>5</sub>), chemical oxygen demand (COD), total suspended solids (TSS), turbidity, nitrate, phosphate, and ammonium was studied. The average removal efficiencies obtained were 95.5% BOD<sub>5</sub>, 94.6% COD, 86.2% TSS, 76.6% turbidity, 82.4% nitrate, 76% phosphate, and 32.9% ammonium. The results show that integrating ABR, HSSFCW, and FCW improves pollutant removal from seed production wastewater, and the treated water can be used for agricultural purposes.

**Key words**: anaerobic baffled reactor, floating constructed wetland, horizontal subsurface flow constructed wetland, performance evaluation, seed production wastewater, wastewater treatment

#### **Highlights**

- The performance of wastewater treatment system in treating seed production wastewater was studied.
- The system integrated anaerobic baffled reactor, horizontal subsurface flow and floating constructed wetlands.
- The system removed COD, BOD5, TSS, turbidity below acceptable limit of Tanzanian national standard for industrial effluent
- The integrated system is promising for pollutant removal from seed production wastewater.

#### **Graphical Abstract**



#### INTRODUCTION

Industrial, municipal, and agricultural wastewaters contribute greatly to water pollution (Shi 1998; Kadirvelu *et al.* 2001; Hagberg 2007). Seed production wastewater is composed mainly of organic

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matter, nutrients, and suspended solids. Discharging such wastewater either untreated or partially treated can cause environmental pollution. Eutrophication can arise from excess loads of nitrogen and phosphorus in aquatic environment (Bu & Xu 2013). Moreover, high organic matter content can cause oxygen depletion, bad odor, and fish kills (Assefa *et al.* 2019). Adopting appropriate wastewater treatment technologies is necessary to minimize pollution.

An anaerobic baffled reactor (ABR) is an anoxic wastewater treatment system with vertical baffles, which the wastewater passes under and over (Bachmann *et al.* 1985). Bacteria move horizontally in the reactor as wastewater passes through, and also tend to rise due to gas production, allowing the wastewater to come into contact with a large active biomass within a short hydraulic retention time (HRT) (Nguyen *et al.* 2010). The design simplicity with its associated short HRT, the ability to sustain high organic loads, and loading shocks are important benefits. ABR is also characterized by low energy consumption and sludge production. Several studies have proved ABR's removal ability for organic matter and suspended solids from wastewaters (Movahedyan *et al.* 2007; Ferraz *et al.* 2009; Alighardashi *et al.* 2015). However, nitrification is restricted in ABRs and the ammonium concentration increases due to the anoxic environment. Post-treatment is needed, therefore, to reduce the concentrations of ammonium, pathogens, and residual chemical oxygen demand (COD), biological oxygen demand (BOD<sub>5</sub>), and total suspended solids (TSS).

Constructed wetlands (CWs) are an effective, efficient, and suitable wastewater treatment system due to their low capital and running cost, simplicity in operation and energy consumption (Njau & Renalda 2010). They are classified on the basis of their hydrology, flow-path, and macrophyte growth forms. There are two types under the hydrologic classification – subsurface flow and surface flow CWs. There are also two types with respect to flow-path – horizontal and vertical flow. In horizontal subsurface flow CWs (HSSFCWs), the wastewater flows horizontally under the bed surface to the outlet zone. Pollutant removal in HSSFCWs is done by physical, chemical, and biological processes including filtration, sedimentation, chemical precipitation, photochemical reactions, photosynthesis, fermentation, nitrification, and denitrification. HSSFCWs show effective removal of BOD<sub>5</sub>, COD, and TSS (Vrhovšek *et al.* 1996; Zhang *et al.* 2009). However, nutrient removal efficiency is considered low in single-stage HSSFCWs (Cottingham *et al.* 1999; Khanijo 2002; Akratos & Tsihrintzis 2006; Rossmann *et al.* 2012).

Floating constructed wetlands (FCWs) are small artificial platforms that allow aquatic plants to grow in water that is typically too deep for them. This allows a unique ecosystem to develop with the potential to capture nutrients and transform common pollutants. The wastewater is treated in the aerobic environment (Tanner *et al.* 2011). In FCWs, the nutrients from wastewater are taken up by plants, while microorganisms in a biofilm formed on the plant roots and mat surface degrade organic matter and provide environment for nitrogen transformation (Shahid *et al.* 2018). FCWs are considered efficient for nutrient removal from eutrophic water bodies (Stefani *et al.* 2011; Bu & Xu 2013; Borne 2014).

Single-stage CWs are not recommended for strong wastewater treatment without having pretreatment. It has been observed that most single-stage CWs had low pollutant removal efficiency in heavily loaded wastewater (Sayadi *et al.* 2012). Study by Wang *et al.* (2014) showed failure of HSSFCWs after use for primary treatment, because of clogging and high pollutant loads in the wastewater.

Subsurface flow CWs accelerate denitrification whereas surface flow CWs accelerate nitrification. In both cases, nitrification or denitrification is limited by the system's anaerobic/aerobic condition. Combining subsurface flow and floating CWs as a final treatment system is expected to reduce the nitrogen components in wastewater through nitrification, denitrification and plant uptake (Saeed *et al.* 2014). Hybrid CWs have been studied for different types of wastewater treatment (El-Khateeb *et al.* 2009; Singh *et al.* 2009; Xiong *et al.* 2011; Ye *et al.* 2012; Saeed *et al.* 2014). However, studies on HSSFCWs combined with FCWs in pollutant removal are limited. Published data are very limited concerning on the use of subsurface flow and FCWs for wastewater treatment from seed production.

The aim of this study was to combine the advantages of HSSFCW and FCW, integrated with ABR, to treat seed production wastewater.

#### **MATERIALS AND METHODS**

#### Study site

The study was conducted at Enza Zaden seed-producing industry in Arusha, Tanzania at  $3^{\circ}24'0.521''$  S latitude and  $36^{\circ}47'16.256''$  E longitude, and 1,192 m above mean sea level. Production comprises vegetable seeds including sweet pepper, paprika, cucumber, and tomato, and wastewater is generated when the seeds are washed. Some 20-30 m<sup>3</sup>/d are generated and stored in a 340 m<sup>3</sup> equalization tank. It is first treated in an ABR before transfer to the HSSFCW and FCW. The system was new and commissioned in June 2020. This study was conducted from June to August 2020, inclusive.

#### **ABR**

The ABR was made up of six compartments with the same cross-sectional area and  $205.3 \text{ m}^3$  total volume. Primary treatment was done in this unit. During the study, the system received  $25 \text{ m}^3$  – wastewater/d from the equalization tank. System dimensions and operating conditions are described in Table 1.

Table 1 | System dimensions and operating conditions

Dimensions	ABR	HSSFCW	FCW
Length (total), m	19.01	19.3	19.12
Length of treatment zone, m	18.75	19	17
Length of inlet and outlet zones, m	0.26	0.3	0.12
Width, m	3.6	8	8
Water depth, m	2	0.5	0.35
Operating conditions			
HRT, days	5	3.8	4.5
$OLR_{range}^{a}$ , kg- $BOD_{5}/m^{3}/d$	0.114-0.174	0.026-0.118	0.011-0.079
$OLR_{average}^{a}$ , kg- $BOD_{5}/m^{3}/d$	0.134	0.068	0.032
OLR <sub>range</sub> , kg-COD/m <sup>3</sup> /d	0.179-0.262	0.049-0.211	0.016-0.2
OLR <sup>a</sup> <sub>average</sub> , kg-COD/m <sup>3</sup> /d	0.208	0.102	0.061

<sup>&</sup>lt;sup>a</sup>Organic Loading Rate.

#### **HSSFCW**

The HSSFCW receives pretreated wastewater from ABR and discharges to the FCW. It was filled with clean aggregate of 12–20 mm diameter and 0.35 average porosity, and planted with the native African aquatic flowering plant *Cyperus alternifolius* (also known as umbrella papyrus) collected from nearby natural wetlands and planted at three rhizomes/m². Above the compacted earth surface, selected sand was used to create a smooth bottom and protect the plastic liner from being torn by rocks etc. The influent flowed horizontally through the gravels and plants to the exit. The HSSFCW cross-section and configuration are shown in Figure 1.

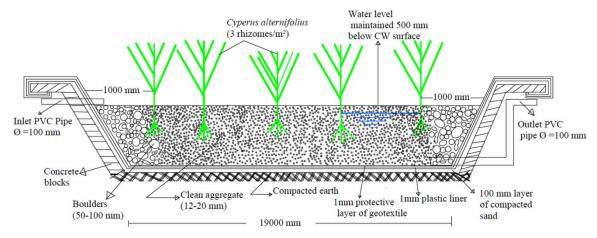


Figure 1 | HSSFCW cross-section.

#### **FCW**

The final stage – the FCW- had four polyethylene foam plate floating mats, each covering 3.75 m<sup>2</sup> and fixed 4 m apart (Figure 2). The mats were covered with *Vetiveria zizanioides* (Vetiver grass). The FCW dimensions and operational conditions are given in Table 1.

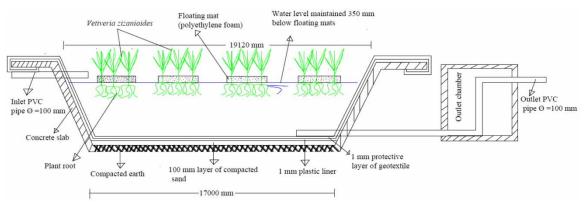


Figure 2 | FCW cross-section.

#### Sampling

Wastewater samples were collected from the inlet and outlet of each system twice weekly by following the APHA recommended standard methods for examination of water and wastewater (APHA 2017) using pre-cleaned 100 ml polyethylene sampling bottles. A total of 108 samples was collected. The bottles were prepared by soaking in 5% HCL overnight and rinsed in the laboratory with distilled water 3–5 times. In the field, the bottles were rinsed 3–5 times with the wastewater to be collected, before sampling. The samples were stored in a cool-box at 4 °C and transported to Nelson Mandela African Institution of Science and Technology laboratories for analysis.

#### Physicochemical analysis

Parameters like pH, temperature, electrical conductivity (EC), and total dissolved solids (TDS) were measured in-situ using a HANNA Multiparameter (HI 9829), also turbidity was analyzed with a Microprocessor Turbidity meter (HI 93703), both instruments are manufactured by HANNA

Instruments Company in Nasfalau, Romania. In addition, the cadmium reduction method was used to determine nitrate, and the ascorbic acid powder pillow method for phosphate, using a HACH DR 2800 spectrophotometer (HACH Company, Berlin, Germany). The Nessler reagent method was used to determine ammonium, while COD was determined by reactor digestion, and  $BOD_5$  by closed manometer.

#### Data analysis

Origin pro version 9.0 (Originlab 2012) and Microsoft Excel were used for data analysis. The pollutant concentration trend and removal efficiency in each treatment unit were obtained. The system's pollutant removal efficiency was calculated using Equation (1).

$$R(\%) = \left(\frac{C_i - C_f}{C_i}\right) * 100 \tag{1}$$

where R is percentage removal efficiency, and  $C_i$  and  $C_f$  the initial and final pollutant concentrations.

#### **RESULTS AND DISCUSSION**

Table 2 shows the average influent and effluent physio-chemical characteristics for each treatment stage:

Table 2 | Physio-chemical characteristics for each treatment stage

	Influent		ABR		HSSFCW		FCW	
Parameter	Average	S.D.	Average	S.D.	Average	S.D.	Average	S.D.
pH	6.8	0.3	6.9	0.2	7.1	0.2	7.53	0.2
Temperature (°C)	23.5	1.6	24.1	1.5	22.9	1.4	21.8	2.1
EC (µs/cm)	1,924	213.5	1,966	241.2	2,034	200.6	2,003	256.6
TDS (mg/L)	962.2	106.8	984.1	120.2	1,017	100.3	1,001.8	128.6

#### **Temperature**

The average temperature tended to increase in transit through influent and ABR effluent and decrease in HSSFCW and FCW effluents (Table 2). The final effluent temperature range was within the Tanzania Bureau of Standards' (TBS) acceptable limit for industrial wastewater effluent (20–35 °C) (TBS 2009). Moreover, the temperatures were within the optimal range for effective biological activity in each stage. Temperature is a key parameter in biological treatment, as it affects the rate of microbial activity (Kadlec & Reddy 2001) – microorganisms in treatment systems generally function effectively in the 20–35 °C range.

#### рН

pH is an important factor in chemical and biological activities. The pH increased from inlet to outlet in each treatment unit (Table 2). In the ABR this might arise from microorganism activity in the anaerobic environment. During anaerobic degradation of carbohydrates or fatty acids in the last process stage, ammonia gas production will lead to an increase in pH. Denitrification (in the HSSFCW)

also increases the pH (Xiong *et al.* 2011). In both HSSFCW and FCW, intensive photosynthesis by submerged, emerged and floating pants also increases the pH (Yin *et al.* 2016). However, the pH in each treatment system was within the optimum range (6.5–8.5) for biological wastewater treatment processes (Metcalf & Eddy 2004), and the pH of the FCW's final effluent was in the range of 7.2–8, within the TBS pH limit (6.5–8.5) for industrial effluent.

#### TDS and EC

TDS and EC increased from inlet to outlet of each unit (Table 2), probably because of pollutant degradation and dissolution of ions (Mtavangu *et al.* 2017). The increase in TDS and EC in ABR might also arise from mineralization, i.e., the conversion of organic carbon into smaller and simpler organic compounds.

#### **TSS**

Figure 3(a) shows the variation of TSS concentration in each treatment stage over time. The respective removal efficiencies were  $47 \pm 8.3\%$ ,  $64.7 \pm 10.2$  and  $28.3 \pm 17.1$  in ABR, HSSFCW and FCW, with  $86.2 \pm 6\%$  performance efficiency for the integrated system. The final effluent from FCW had an average concentration of  $51.44 \pm 23$  mg-TSS/L and met TBS' standard for industrial effluents.

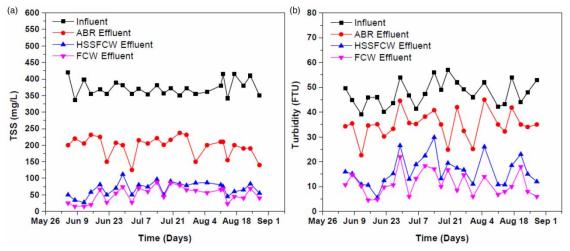


Figure 3 | TSS and turbidity concentration over time - influent, ABR, HSSFCW and FCW.

#### **Turbidity**

Figure 3(b) presents turbidity concentration variation across three treatment units. Average turbidity removal efficiency was thus  $26.6 \pm 9.9\%$ ,  $53.5 \pm 14.2$ , and  $31 \pm 16.2$  in the ABR, HSSFCE and FCW respectively. The integrated system's turbidity removal efficiency was  $76.6 \pm 9.5\%$ , and the final effluent reported  $11.2 \pm 4.8$  FTU, within TBS' maximum permissible limit.

#### Nitrogen and organic species

Table 3 shows the average pollutant concentration in each treatment unit. Based on BOD<sub>5</sub>, COD,  $NO_3^-$ ,  $NH_4^+$  and  $PO_4^{3-}$  content in the raw wastewater it is classified as high strength wastewater

Table 3 | Pollutant concentration at different treatment stages

		Raw		ABR		HSSFCW		FCW	
Parameter	Unit	Average	S.D.	Average	S.D.	Average	S.D.	Average	S.D.
BOD <sub>5</sub>	mg/L	688.8	95.7	206.0	81.4	59.4	35.2	26	12.4
COD	mg/L	1,074	130.5	301.6	135	107.7	83	58.3	39.7
$NO_3^-$	$mg-NO_3^-/L$	376.9	87.5	332.9	86.5	173.8	49.1	66.3	25.8
$NH_4^+$	$mg-NH_4^+/L$	123.6	18.4	141.5	18	122.3	15.4	106.3	18.7
$PO_4^{3-}$	$mg-PO_4^{3-}/L$	60.2	11.5	52.9	10.3	29.7	8.7	14.2	5.8

(Metcalf and Eddy 2004). Furthermore, the BOD<sub>5</sub>/COD ratio was between 0.6 and 0.8, indicating that it is highly biodegradable (Zaher & Hammam 2014).

The removal efficiencies of the treatment stages and their pollutant removal performance over time are presented in Table 4 and Figure 4 respectively. In the ABR, microorganisms degrade organic matter to methane and carbon dioxide (Dinsdale *et al.* 2007). COD removal efficiency in the ABR ranged from 31.2% to 88.5%. At the beginning of the study, ABR removal efficiency was below 50% (similar for  $BOD_5$ ) but, after one month of operation, efficiency began to increase, and reached 88.5% at the end of the second month. The average removal efficiency during high level performance (second and third month) was  $81 \pm 10.1\%$ . The ABR's COD removal efficiency in this study was similar to that in other studies, for example that by Ferraz *et al.* (2009) on cassava wastewater treatment with 83% COD removal efficiency reported for 3.5 HRT and 2 g-COD/ L/d OLR. Minh & Phuoc (2014) studied ABR performance in domestic wastewater treatment and obtained 72–74% COD removal at OLRs of 1.5–2.7 kg-COD/m³/d and 3 hours HRT.

Table 4 | Pollutant removal efficiency by treatment unit and integrated system (units as in Table 3)

	ABR		HSSFCW		FCW		OVERALL	
Parameter	Average	S.D.	Average	S.D.	Average	S.D.	Average	S.D.
BOD <sub>5</sub>	70.6	11.7	71.1	10.6	42.5	20.1	95.5	1.9
COD	71.6	13.6	65.7	13.4	40.9	19.7	94.6	4
$NO_3^-$	12	7.4	46.8	11.9	61.5	11.7	82.3	6
$\mathrm{NH_4^+}$	-15.3	11.1	13.2	8.6	32.9	6.5	32.9	13.1
$PO_4^{3-}$	11.9	8.5	43.7	12.4	52.9	12.5	76	10.5

The ABR's influent and effluent had BOD<sub>5</sub> concentrations ranging from 591 to 900 mg/L and 80 to 360 mg/L, respectively with the removal efficiency ranged from 44.9 to 91.1%. After the first month, ABR BOD<sub>5</sub> removal performance was higher than that reported by Mahenge & Malabeja (2018) (82%) for municipal wastewater treatment in Tanzania, which had an average influent BOD<sub>5</sub> concentration of 314 mg/L.

Denitrification –  $NO_3^-$  reduction – was observed in the ABR, which removed  $NO_3^-$  as nitrogen gas (Stuckey & Barber 2000). Nitrate removal efficiency was low ( $12 \pm 7.4\%$ ), however, compared to HSSFCW ( $46.8 \pm 11.9\%$ ) and FCW ( $61.5 \pm 11.7\%$ ), which might be attributable to limited organic carbon availability because of organic matter oxidation in the system. The  $NH_4^+$  also increased in transit through the ABR because it was released during the anaerobic degradation of organic matter in the anaerobic environment (Hahn & Figueroa 2015; Mahenge & Malabeja 2018). Moreover,  $NO_3^-$  reduction in anoxic environments also leads to the formation of  $NH_4^+$  (Semba *et al.* 2020).

Table 4 shows the average removal efficiencies for BOD<sub>5</sub>, COD, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup> and PO<sub>4</sub><sup>3-</sup> in HSSFCW. Microorganisms attached to the plant roots and rhizomes, and on the substrate (gravel), degrade the

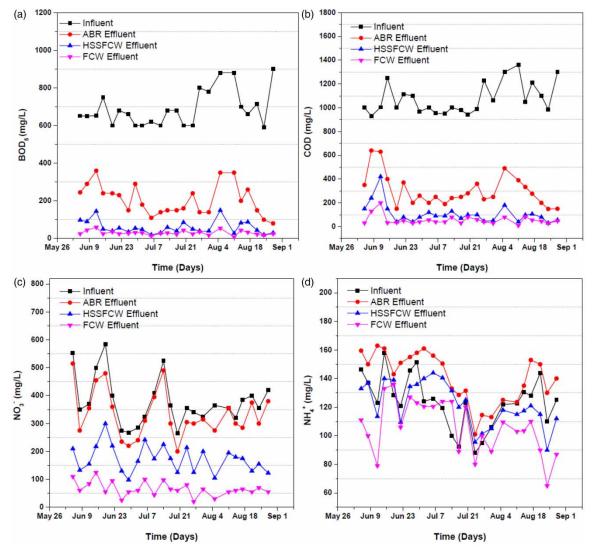


Figure 4 | Pollutant concentrations by treatment unit – (a) BOD<sub>5</sub> (b) COD (c) NO<sub>3</sub> (d) NH<sub>4</sub>.

organic matter. In this study, the ammonium removal efficiency was lower than that of  $BOD_5$  and COD because the organic removal and biological nitrification pathways conflict (Saeed *et al.* 2014). When organic matter degradation was high, the oxygen was depleted, inhibiting nitrification. The nitrate form of the nitrogen component, however, was removed by denitrification and plant uptake. Moreover, phosphate was removed by sedimentation, filtration, precipitation, and a small amount plant uptake.

In FCW, organic matter was removed by microorganisms attached to the floating mat and the plant roots. Because the environment is oxic, nitrification was not limited. Ammoniacal species were converted to nitrite and nitrate, which were then available for uptake by floating plants. Phosphorous was also removed in this stage by sorption, physical entrapment in the root zone, and plant uptake.

#### **Integrated system performance**

Figure 4 shows the pollutant removal performance of each stage over time. The integrated system's average  $BOD_5$  removal efficiency was  $95.5 \pm 1.9\%$  and the final effluent  $BOD_5$  concentration from FCW was below TBS standard for industrial effluent discharge (Table 5).

The combined system was designed for optimal nutrient removal. The highest nitrification rate was observed following the final treatment stage because of the aerobic conditions in the FCW. The final

Table 5 | Pollutant concentrations at the system inlet and outlet, and the Tanzanian national discharge standards

Parameter	Unit	ABR inlet	FCW outlet	TBS discharge values
pН	_	$6.8\pm0.3$	$7.5\pm0.2$	6.5-8.5
TDS	mg/L	$962.2 \pm 106.8$	$1{,}001.8 \pm 128.6$	-
EC	μs/cm	$1,924 \pm 213.5$	$2{,}003.3\pm256.63$	-
Temperature	°C	$23.5\pm1.6$	$21.8\pm2.1$	20–35
TSS	mg/L	$373 \pm 23.77$	$51.44 \pm 23$	100
Turbidity	FTU	$47.7\pm5$	$11.2 \pm 4.8$	300
$BOD_5$	mg/L	$688.8 \pm 95.7$	$26\pm12.4$	30
COD	mg/L	$1,074\pm130.5$	$58.3 \pm 39.7$	60
$NO_3^-$	$mg-NO_3^-/L$	$376.9 \pm 85.5$	$66.3 \pm 25.8$	20
$\mathrm{NH_4^+}$	$mg-NH_4^+/L$	$123.6 \pm 18.4$	$106.3 \pm 18.7$	-
$PO_4^{3-}$	$mg-PO_4^{3-}/L$	$60.2\pm11.5$	$14.2\pm5.8$	_

effluent NO<sub>3</sub><sup>-</sup> concentration exceeded TBS' permissible discharge level (Table 5). The high nitrate concentration in the final effluent might arise because of its high initial concentration in the influent and low levels of denitrification in the ABR and HSSFCW (Assefa *et al.* 2019). There was a large input of nitrate at the influent as shown in the inlet values of ABR, which arose because the industry discharges excess artificial fertilizer (used to grow different vegetables for seed extraction process) from the greenhouses to the equalization tank. This information about artificial fertilizer discharge was not provided to the designers. As the ABR was sized mainly for the removal of organic matter, the size prescribed may not be adequate for denitrification. Denitrification could be enhanced in the ABR by supplementary carbon addition, perhaps as methanol, sugar, volatile fatty acids, etc (Assefa *et al.* 2019).

Ammonium increased in the ABR due to the anaerobic transformation of organic nitrogen to ammonium and, possibly, also through nitrate reduction to ammonium. It decreased in the HSSFCW and FCW stages, however, because both are oxic and enhance nitrification process. It is well known that HSSFCW has oxic areas around the root zone due to pumping of oxygen from the leaves through the stem. The average  $NH_4^+$  removal efficiency was low (Table 5) and is thought to arise due to the breakdown of organic nitrogen in the ABR (anaerobic) to produce  $NH_4^+$ .

Figure 5 shows the variation of phosphate concentration in each treatment stage. The integrated system achieved  $76 \pm 10.5\%$  average phosphate removal efficiency. Both HSSFCW and FCW played important roles in this.

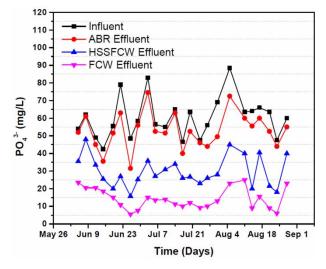


Figure 5 | Variation in phosphate concentration by stage through time.

In this study, the integrated system's performance in BOD<sub>5</sub> and COD removal was better than that in El-Khateeb *et al.*'s study (2009). They used an integrated system comprising an up-flow anaerobic sludge blanket reactor, and free water-surface and subsurface flow CWs. The integrated system discussed here also showed better removal efficiency for TSS, BOD<sub>5</sub>, COD, and phosphate than reported by Singh *et al.* (2009), who used an integrated system comprising ABR, HSSFCW, and vertical subsurface flow CW treating strong municipal wastewater.

#### **CONCLUSIONS**

Combining different wastewater treatment technologies improve pollutant removal efficiency from wastewater. The performance of HSSFCW integrated with FCW and ABR to treat seed industrial wastewater was evaluated in this study. The removal rates of TSS, turbidity, COD, BOD<sub>5</sub>,  $NO_3^-$ ,  $NH_4^+$  and  $PO_4^{3-}$  were all good. The pollutant concentrations in the effluent from the last treatment stage were below TBS' permissible maximum for industrial effluent except nitrate.

The study's results indicate that using an integrated treatment series consisting of ABR, HSSFCW, and FCW is promising for pollutant removal from seed production wastewater. The treated wastewater has potential for use in irrigation.

#### **DATA AVAILABILITY STATEMENT**

All relevant data are included in the paper or its Supplementary Information.

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### **Uncorrected Proof**

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