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Removal of COD from synthetic wastewater in vertical flow constructed wetland

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Received 26 March 2019; Revised 17 June 2019; Accepted 19 June 2019

Guizhou Provincial Science and Technology Plan Project, Grant/Award Number: QIANJIAOHELHZI[2016]7283; Guangdong College Wastewater Ecological Treatment and Water Rehabilitation Engineering Technology Research Center Construction Project of Guangdong Provincial Department of Education, Grant/Award Number: 2012gczxA1004; Anshun College Ph.D. Fund Project, Grant/Award Number: (asubsjj2016) No. 07; National Natural Science Foundation of China, Grant/Award Number: 41271245; Guizhou Provincial Support Program for Top Talents in Science and Technology in Colleges, Grant/Award Number: QIANJIAOHEKY[2016]097

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DOI: 10.1002/wer.1168

Abstract

Three vertical flow constructed wetlands, that is, system A (planted with *Pennisetum sinese* Roxb), system B (planted with *Pennisetum purpureum* Schum.), and system C (without plants as the control) were constructed to estimate the contribution of substrates, plants, and microorganisms to organic matter removal. The organic compounds accumulated in the substrate in systems A, B, and C were 12.03%, 11.91%, and 9.4%, respectively. Synergistic utilization of organic compounds by microorganisms and plants in systems A, B, and C were 80.95%, 81.58%, and 80.11%, respectively. Substrate interception and adsorption of organic compounds in plant systems A and B were more extensive than in the nonplant control system C. The total accumulative and absorptive capacity of systems A, B, and C was as follows: B (2,713 g) > A (2,698 g) > C (2,076 g). The amounts of insoluble organic accumulated on the upper substrates of the three systems showed the order C > A > B. No constructed wetland clogging occurred for A and B systems during the experiment, while system C suffered clogging in early September. Therefore, substrate blockage may be related to the type of organic compound accumulated. Accumulation of insoluble organic matter is the direct cause of system blockage.

Practitioner points

- Substrate interception and adsorption of organic compounds in plant systems were more extensive than those in the nonplant control system.
- Distribution characteristics of the surface layer were significantly higher than those of the middle and bottom layers.
- Substrate blockage is related to the type of organic compound accumulated.
- Accumulation of insoluble organic compounds may be the direct cause of system blockage.
- The upper substrate is the main site for organic compound removal.

Key words

blockage; insoluble organic compound; organic compound; vertical flow constructed wetlands

Introduction

Constructed wetlands (CW) are an emerging wastewater treatment technology with low investment, low energy consumption, and convenient management (Babatunde, Zhao, & Zhao, 2010; Huang, Reneau, & Hagedom, 2000; Machate, Noll, Behrens, & Kettrup, 1997; Ranieri, Gorgoglione, & Solimeno, 2013; Srinivasan, Weaver, Lesikar, & Persyn, 2000; Tang, Yu, Zhou, Zhang, & Li, 2017; Vymazal, Greenway, Tonderski, Brix, & Mander, 2006). They are considered a promising ecological treatment technology (Cui, Ouyang, Gu, Yang, & Xu, 2013; Kadlec & Knight, 1996). In recent years, the economic and environmental benefits of constructed wetlands have been recognized by several scholars (He, Peng, Hua, Zhao, & Xiao, 2018; Lin et al., 2005; Sindilariu, Brinker, & Reiter, 2009; Sindilariu, Wolter, & Reiter, 2008). Vertical flow constructed wetlands show good performance in removing nitrogen, phosphorus, and organic compounds due to their small area and excellent oxygen transfer characteristics (Cooper, 2005; Tang, Huang,

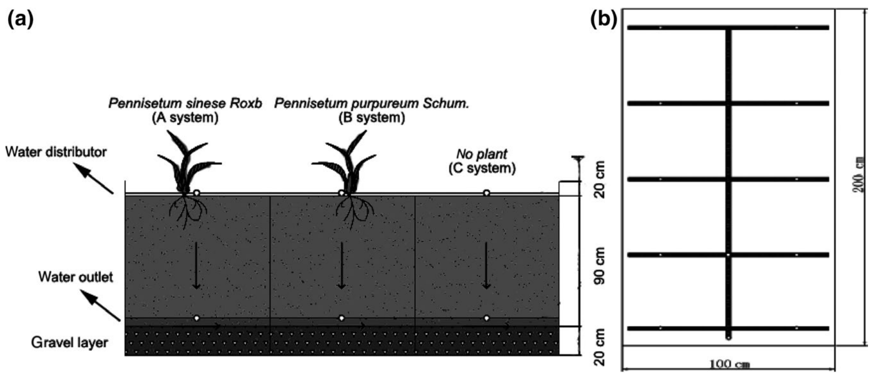

Figure 1. Schematic diagram of the simulated vertical flow constructed wetland (a) and surface water distributor (b).

Scholz, & Li, 2011; Zhou, Gao, Zhang, & Wu, 2018). Because of its obvious advantages in wastewater purification, this system type has been widely used in China (Du, Xu, Li, & Zheng, 2014). The factors affecting the removal of pollution from constructed wetlands mainly include substrates, plants, and microorganisms (Cui et al., 2013; Shalla, Kinross, Read, & Marland, 2000; Xu, Huang, Wang, & Cui, 2015). While there is considerable research with regard to organic matter treatment processes and performance associated with CW systems, many studies have not quantified the contribution of plants, substrates, and microorganisms to the removal of organic matter (Korkusuz, Beklioğlu, & Demirer, 2005; Xu, Chen, Huang, Cui, & Wang, 2016), and the broad range of results and interpretations has led to a disappointing lack of consensus that may be limiting wider application of CW as a treatment option in practice (Collison & Grismer, 2013). Since the composition of organic pollutants in wastewater is usually complicated, measuring the content of various organic substances is quite difficult. In this regard, BOD, COD, TOC, and TOD are generally used as indicators in actual work. Given continuous improvements in living standards, the increase in COD emissions from domestic wastewater has become the main reason behind the increase in total COD emissions in China. The main objectives of this paper are as follows: (a) to explore the main factors affecting organic matter degradation in vertical flow constructed wetlands; (b) to estimate the contribution of substrate adsorption retention and plant microbial absorption and decomposition to this degradation and quantify the contribution of substrates, plants, and microorganisms to organic matter removal in vertical constructed wetland system.

Materials and Methods

Construction of the vertical flow constructed wetlands

There were three vertical flow constructed wetland systems with a coagulation cement structure constructed, which are system A (planted with *Pennisetum sinese* Roxb), system B (planted with *Pennisetum purpureum* Schum.), and system C (planted nothing). The vegetation types of *P. sinese* Roxb and *P. purpureum* Schum. were transplanted into systems A and B, respectively, with a density of 10 plant/m^2. The length, width, and height of the wetlands were 200, 100, and 130 cm, respectively. The wetland was filled with a 20-cm-thick layer of gravel with the diameter of 20 mm in the bottom to avoid clogging, and 90 cm of river sand of 1–2 mm is the substrate layer; a 20 cm water distribution area was set aside in the upper portion of the wetland. A PVC pipe with a 5 cm diameter was used as the water distribution system. To ensure uniform distribution of water in the system, water distribution holes were set every 4 cm on the PVC pipe, and the diameter of the hole was 1 cm. At the same time, a set of catchment systems was installed at the bottom of the wetland system. A schematic of the constructed wetlands is shown in Figure 1.

Quality and load of inlet water

The inlet water was prepared using chemically pure reagents to simulate domestic wastewater: 400 L tap water, 70 g milk powder, 60 g soluble starch, 18 g urea, 10 g $(NH_4)_2SO_4$, 6 g $MgSO_4$, 6 gKH_2PO_4, 60 g $NaHCO_3$, and 50 ml 1% $FeCl_3$. The water quality indices are pH 6.39–6.7, TN: 30–50 mg/L, TP: 3–5 mg/L, COD: 235–359 mg/L, and DO: 4–6 mg/L.

Experimental process

The vertical flow constructed wetland was constructed on March 10, and operation began on March 28 and ended on September 26. The system hydraulic load was 20 cm/day. The method of wastewater feeding was as follows: From March 28, CWs were pumped continuously with simulated wastewater for 8 hr every day, and each system was irrigated 400 L/day. From April 11, water samples were collected from the bottom outlet and analyzed once a week. The aboveground biomass of CW plants was harvested once a month. At the end of the experiment, five samples were collected individually from the upper layer (0–30 cm), middle layer (30–60 cm), and lower layer (60–90 cm) of each constructed wetland. These five samples were mixed to obtain one representative sample for each layer.

Analytical methods

Determination of different organic substances—total organic compounds (TCOD)—were determined by the standard potassium dichromate method (National Environmental Protection Agency, 2002), and dissolved organic compounds

Table 1. Different organics content in wastewater

CONCENTRATION	DISSOLVED ORGANIC COMPOUND (DOC)	PARTICULATE ORGANIC COMPOUND (POC)	TOTAL ORGANIC COMPOUND (TOC)
Concentration range (mg/L)	200–240	100–160	300–400
Mean	220	130	350
Organic compound content (g)	13,200	7,800	21,000

(DOC) were determined by filtering wastewater through a 0.45-μm filter paper. Particulate organic compounds (POC) were calculated as TCOD–DOC (Shi, Hu, Zhou, & Chen, 2011).

Determination of soluble organic matter and dissolved organic matter content: mass of soluble organic matter = mass of soluble solid − mass of soluble solid after burning; mass of insoluble organic matter = total solid mass − mass after total solid burning - mass of soluble organic matter (Fu, Ye, Gu, & He, 2010).

Detection of the amount of organic compounds intercepted between substrates—exactly 5 ml of a substrate sample—was taken out and washed with 200 ml of distilled water. Then, the organic compound content of substrate interception in aqueous solution was determined by the filtration membrane method. The calculation method was as follows: The organic compound intercepted by substrate = total solid weight (dried at 103–105°C) − total solid weight after burning (600°C) (Du, Xu, & Wang, 2010).

Detection of organic compounds absorbed by the substrate: The ultrasonic chemical exfoliation method was used to remove the substrate biofilm, which was then dried, weighed (W_1), and, then, burned in a muffle furnace for 60 min to a constant weight (W_2). The organic content absorbed by the substrate was calculated as $W_2 - W_1$ (Du et al., 2010).

Statistical method

Excel 2007 and SPSS 17.0 software were used to analyze the variance of the data. Mean values and standard errors of correlation analysis were calculated. The Duncan method was used to analyze and compare the multiple comparisons. Because of the large scale of wetlands, no treatment duplication was set up, and the statistical analysis duplication in this paper is the absolute duplication of sampling.

Results

Different types of organic compound in wastewater

When evaluating organic compounds with COD, understanding the basic characteristics of organic compounds is necessary because it includes nonbiodegradable and biodegradable organic compounds; that is, soluble and granular COD concentrations. The types of organic compounds in wastewater treated by vertical flow wetland system are total organic compounds (TCOD), particulate organic matter (POC), and dissolved organic compounds (DOC). Based on the inlet organic load of the vertical flow constructed wetland over 5 months (actual inlet, 150 days), the average amount of POC, DOC, and TCOD in the water of the vertical flow constructed wetland was 7,800, 13,200, and 21,000 g, respectively (Table 1). The proportion of POC in the water was 37.1%.

Interception and adsorption of different organic substances by the substrate

Table 2 demonstrates that the substrates of CW can absorb and intercept different types of organic compounds. In the three systems, the content of soluble organic matter, interception organic matter and adsorption organic matter in the substrate layer of system A and system B were significantly

Table 2. Organic matters accumulation in different depth of substrate (g/ml)

SYSTEM	DEPTH (CM)	SOLUBLE ORGANIC COMPOUND (G/ML)	INSOLUBLE ORGANIC COMPOUND (G/ML)	ORGANIC COMPOUNDS INTERCEPTED BY THE SUBSTRATE (G/ML)	ORGANIC COMPOUNDS ADSORBED BY THE SUBSTRATE (G/ML)
A	0–30	0.00069	0.00134	0.00203	0.000157
	30–60	0.00035	0.00063	0.00098	0.000119
	60–90	0.00039	0.00081	0.0012	0.00011
B	0–30	0.00058	0.00114	0.00172	0.000176
	30–60	0.00053	0.00064	0.00117	0.000106
	60–90	0.0004	0.00088	0.00128	0.00007
C	0–30	0.00013	0.00153	0.00166	0.00009
	30–60	0.00023	0.00051	0.00074	0.00006
	60–90	0.00024	0.00065	0.00089	0.00002

Note. Because of the large scale of the experimental wetland system, repetitive experiments could not be performed. All experimental data are sample repetition, and values indicated in the table are the average values of three samples repetition.

Table 3. The correlation analysis of different organic matter and the COD removal efficiency

	COD	SOLUBLE ORGANIC COMPOUND	INSOLUBLE ORGANIC COMPOUND	INTERCEPTED ORGANIC COMPOUND	ADSORBED ORGANIC COMPOUND
COD (%)	1				
Soluble organic compound (g/cm^3)	0.052	1			
Insoluble organic compound (g/cm^3)	−0.732*	−0.249	1		
Intercepted organic compound (g/cm^3)	−0.602	0.529	0.690*	1	
adsorbed organic compound (g/cm^3)	−0.247	0.100	0.666	0.658	1

Note. *In 0.05 level (double side).

higher than that in the substrate layer of system C. However, the content of insoluble organic matter in system C was higher than that in system A and system B. In the present experiment, by September 10 or so, a backwater blockage was observed in system C but not in systems A and B. Zhan, Wu, and Xu (2003) proposed that accumulation of organic compounds is the main cause of wetland blockage. Table 2 reveals that the content of insoluble organic matter in the upper layer of system C (0–30 cm) is obviously higher than that in upper layers of systems A and B. In this experiment, a backwater blockage was observed in system C, which indicates that accumulation of insoluble organic matter may be an important factor contributing to the system blockage. This conclusion is similar to the research results of Richardson and Vepraskas (2000). Kadlec and Knight (1996) also pointed out that, in horizontal subsurface flow constructed wetlands, solid interception of large insoluble particles is the main mechanism of suspended compound removal and mainly occurs in the first few meters of wetland (Wang, You, & Yuan, 2014). When wetlands are operated, suspended particulates and refractory organic compounds are intercepted in large quantities, and the pores between fillers decrease continuously, resulting in blockage.

Relationship between different organic compounds and COD removal

Table 3 shows that insoluble organic matter and substrate interception of organic compound present a positive correlation at

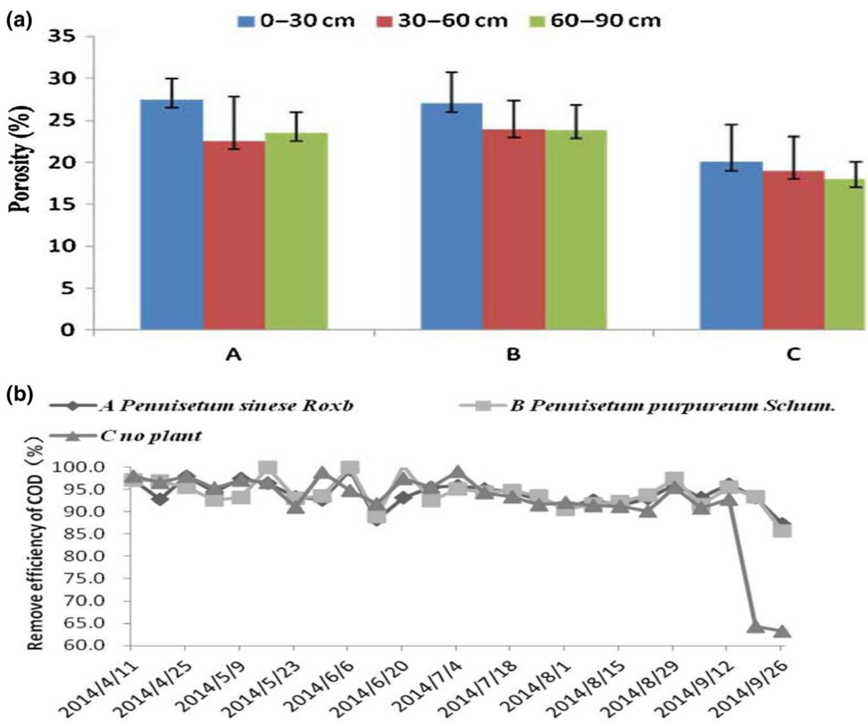

Figure 2. (a) Porosity in the different layers and (b) Removal efficiency of COD in three vertical flow constructed wetlands.

the 0.05 level (R = 0.690*). If the organic compound accumulates in the substrate, it cannot be decomposed in time and reduces the porosity of the substrate; thus, it causes a blockage and reduces the treatment effect (Fu et al., 2010; Tanner & Sukias, 1995). Through correlation analysis of the accumulation of organic compounds and COD removal, insoluble organic matter is an important source of organic compounds intercepted by the substrate and has a significant negative correlation with COD removal efficiency at the 0.05 level (R = −0.732*). In the later stage of the experiment, a large area of water accumulation occurred in system C, indicating obvious blockage occurrence, and a reduction in porosity validity (Figure 2). The removal rate of COD in system C decreased significantly. This finding indicates that accumulation of insoluble organic matter in the wetland will reduce the COD removal rate, possibly because an accumulation of insoluble organic matter in the substrate prevents microorganisms from being decomposed in time, resulting in formation and accumulation of sludge, blockage of substrate voids, and reduction in porosity validity. The formation of a substrate blockage results in a decrease in the wastewater treatment rate. This result is similar to the results of Tong, Zhu, and Ruan (2007), and the amount of COD degradation is related to the effective pore volume of the filler (Xu et al., 2016). Removal of pollutants by wetland substrates may eventually become saturated, and the removal efficiency of pollutants may decrease when the adsorption threshold is exceeded. Therefore, decontamination of the substrate in the constructed wetland involves interception of the contaminants in the bed as a filter medium rich in pores, on the one hand, by plant absorption and microbial decomposition and, on the other hand, by regular replacement of the filler during the operation of the constructed wetland (Ibekwe, Lyon, Leddy, & Jacobson-Meyers, 2007; Zhang, Xu, He, Zhang, & Wu, 2012). During the initial operation of constructed wetlands, the effective porosity of the substrate was high, and its interception and adsorption effects were strong. Over the long-term operation of the constructed wetland, however, the adsorption tendency of the substrate was saturated, and the adsorption of pollutants by the substrate can often be neglected (Wu, Xu, Zhang, He, & Wu, 2015).

Estimation of organic compound accumulation in different forms of wetland substrate

Organic compound accumulation between substrates in the constructed wetlands consists of two parts: soluble organic and insoluble organic (Fu et al., 2010). Between these types, soluble organic compounds are easier to decompose and utilize by microorganisms within a short time. The accumulation of organic compounds between substrates with different vertical directions and heights is shown in Table 4. According to Table 4, the organic compound accumulated between substrates was mainly insoluble organic, and the accumulation of an insoluble organic compound in the upper 0–30 cm substrate layer was significantly higher than that in the two other layers. Perhaps soluble organic matter is easily absorbed and utilized by microorganisms and plants; on the contrary, insoluble organic matter is more difficult to be utilized (Shi et al., 2011).

Changes of organic substrate absorption

The amount of organic compounds absorbed by the substrate in the vertical direction of the different wetland systems is shown in Table 5. Adsorption of organic compounds on the substrate was mainly concentrated in the 0- to 30-cm substrate layer. The substrate adsorption capacity of the plant systems was significantly higher than that of the nonplant system. In fact, the substrate adsorption capacity of system B with plants was about 109 g higher than that of nonplant system C, which indicates that plants can effectively increase the adsorption capacity of the substrate for organic compounds. When studying the treatment of dairy wastewater by subsurface constructed wetlands

Table 4. Average quantity of different organic matters accumulation of substrate in wetland

SYSTEMS	DEPTH (CM)	VOLUME OF SUBSTRATE LAYER (M^3)	SOLUBLE ORGANIC COMPOUND (G/M^3)	SOLUBLE ORGANIC COMPOUND (G)	INSOLUBLE ORGANIC COMPOUND (G/M^3)	INSOLUBLE ORGANIC COMPOUND (G)
A	0–30	0.6	690	414	1,340	804
	30–60	0.6	350	210	630	378
	60–90	0.6	390	234	810	486
	Total	1.8	1,430	858	2,780	1,668
B	0–30	0.6	580	348	1,140	684
	30–60	0.6	530	318	640	384
	60–90	0.6	400	240	880	528
	Total	1.8	1,510	906	2,660	1,596
C	0–30	0.6	130	78	1,530	918
	30–60	0.6	230	138	510	306
	60–90	0.6	240	144	650	390
	Total	1.8	600	360	2,690	1,614

Note. Because of the large scale of the experimental wetland system, repetitive experiments could not be performed. All experimental data are sample repetition, and values indicated in the table are the average values of three samples repetition.

with and without vegetation, Tanner and Sukias (1995) found that the average organic matter accumulation in vegetated wetlands is 0.4–2.3 kg/m^2 per year, while that in vegetated wetlands was close to 4 kg/m^2 per year.

Changes of organic substrate interception

Organic substrate interception by the vertical substrate of the different wetland systems is shown in Table 6. Interception of organic compounds by the substrate was mainly concentrated on the upper 0- to 30-cm layer, and some of them were even two times higher than the accumulation of organic compound on the middle and bottom layers. This finding is similar to the research results of Tanner and Sukias (1995), who found that accumulation of organic compounds within 10 cm of the surface was 2–8 times that below 10 cm. In the present experiment, the substrate accumulation of the three systems showed the order A (2,526 g) > B (2,502 g) > C (1,974 g), and large root systems were beneficial to the interception of organic compounds. However, substrate accumulation differed among the root types of systems A and B, which indirectly proves the importance of choosing wetland plants. This result suggests that plants in wetlands can improve the removal efficiency of organic compounds by the substrate, possibly because the large root system of plants increases the effective porosity of the substrate, which is conducive to the adsorption and interception of organic compounds in wastewater and improves the removal efficiency of these compounds.

Estimation of organic migration and transformation

The removal of organic matter in vertical flow constructed wetlands mainly includes four aspects, and the estimation formula can be expressed as follows (Fu et al., 2010):

$$OM_{total} = OM_{ad} + OM_{in} + OM_{p\text{-}m} =$$
$$\text{TCOD} - \text{Amount of organic compounds flowing out with water}$$

In the formula, OM_{total} denotes total organic compound removal from the wetland system (g); OM_{ad}—organic compound adsorbed by the substrate (g); OM_{in}—organic compound intercepted by the substrate (g); $OM_{p\text{-}m}$—organic compound from synergistic plant–microorganism absorption and utilization (g); TCOD represents the total amount of organic matter in wastewater (g).

The amount of organic compounds flowing out with water = total organic compound (g) × average COD removal rate of the system (%).

Table 7 illustrates that most of the organic compounds entering the wetland system can be removed by plant microorganisms through decomposition. The amount of organic compounds intercepted between substrates is larger than that adsorbed by the substrate, but the effect of the substrate is smaller than that of plants and microorganisms. The removal pathways and rates of the different treatment systems differed. The removal of organic compounds by the substrate was relatively high in plant systems A and B; in fact, little difference was observed between the two wetland plants. This result indicates that wetland plants can improve the effect of the substrate on pollutant removal, which may be due to the mutually beneficial synergy of microorganisms and plants in the combined plant–microorganism system, yielding a complex soil–plant–microorganism system to degrade pollutants together. Plants provide a place of activity for microorganisms through roots (Cheng, Li, & Wang, 2005), and microbes in plants can increase the resistance of plants to external coercion, such as extreme temperature, water, nutrient conditions, and pathogen infestation, thereby enhancing plant degradation of pollutants. A plant–microbe combination system can promote the rapid degradation and mineralization of pollutants (Fu, Xie, & Dai, 2017). Biological action is generally believed to be the core factor of wastewater purification in constructed wetlands (Liang, Wu, Cheng, Zhou, & Hu, 2003). Soil microorganisms play an important role in pollutant removal in wetlands (Stottmeister et al., 2003). The contribution of the plant–microorganism combination in the three systems to COD removal can reach 80%, which directly proves the importance of microorganisms in pollutant degradation.

Conclusions

1. Effect of substrates on COD removal: wetland plants are beneficial to the interception and removal of organic compounds. The accumulation of organic matter occurs mainly in the upper substrate (0–30 cm).

Table 5. Average absorption quantity of organic matter for substrate in wetland

		A		B		C	
SUBSTRATE LAYER (CM)	VOLUME OF SUBSTRATE LAYER (M^3)	TOTAL ORGANIC MATTER ADSORPTION (G/M^3)	TOTAL ORGANIC MATTER ADSORPTION (G)	TOTAL ORGANIC MATTER ADSORPTION (G/M^3)	TOTAL ORGANIC MATTER ADSORPTION (G)	TOTAL ORGANIC MATTER ADSORPTION (G/M^3)	TOTAL ORGANIC MATTER ADSORPTION (G)
0–30	0.6	157	94.2	176	105.6	90	54
30–60	0.6	119	71.2	106	63.6	60	36
60–90	0.6	11	6.6	70	42	20	12
Total	1.8	287	172	352	211.2	170	102

Table 6. Average quantity of organic matter accumulation of substrate in wetland

SUBSTRATE LAYER (CM)	VOLUME OF SUBSTRATE LAYER (M^3)	A		B		C	
		TOTAL ORGANIC COMPOUND ACCUMULATION (G/M^3)	TOTAL ORGANIC COMPOUND ACCUMULATION (G)	TOTAL ORGANIC COMPOUND ACCUMULATION (G/M^3)	TOTAL ORGANIC COMPOUND ACCUMULATION (G)	TOTAL ORGANIC COMPOUND ACCUMULATION (G/M^3)	TOTAL ORGANIC COMPOUND ACCUMULATION (G)
0–30	0.6	2,030	1,218	1,720	1,032	1,660	996
30–60	0.6	980	588	1,170	702	740	444
60–90	0.6	1,200	720	1,280	768	890	534
Total	1.8	4,210	2,526	4,170	2,502	3,290	1,974

Table 7. Estimated average organic matter migration of substrate in wetland

ORGANIC COMPOUND MIGRATION PATHWAY	A		B		C	
	NUMERICAL VALUE	PERCENTAGE	NUMERICAL VALUE	PERCENTAGE	NUMERICAL VALUE	PERCENTAGE
Organic compound absorbed by the substrate	172	0.819	211	1.005	102	0.486
Organic compound accumulated by the substrate	2,526	12.03	2,502	11.91	1,974	9.4
Organic compound released with water	1,302	6.2	1,155	5.5	2,100	10
Amount of plants and microorganisms absorbed and utilized	17,000	80.95	17,132	81.58	16,824	80.11

This result indicates that the upper substrate in vertical flow constructed wetlands is the main site for organic compound accumulation.

2. Degradation rules of COD: Microbial degradation played a major role in COD degradation. The effect of microbial degradation exceeded 80% in all three systems.
3. Wetland blockage: The total accumulative adsorptive capacity of systems A, B, and C in estimates of substrate accumulation is as follows: B (2,713 g) > A (2,698 g) > C (2,076 g). In estimates of insoluble organic, the amounts of insoluble organic accumulated on the upper substrates of the three systems showed the order C (1,530 g/m^3) > A (1,340 g/m^3) > B (1,140 g/m^3). However, only System C suffered clogging in early September, which indicates that substrate blockage may be related to the type of organic compound accumulated. Accumulation of insoluble organic matter is the direct cause of system blockage.

Acknowledgments

This work is funded by the Guizhou Provincial Support Program for Top Talents in Science and Technology in Colleges (QIANJIAOHEKY[2016]097), Guizhou Provincial Science and Technology Plan Project (QIANJIAOHELHZI[2016]7283), Anshun College Ph.D. Fund Project (asubsjj2016) No. 07, National Natural Science Foundation of China (41271245) and Guangdong College Wastewater Ecological Treatment and Water Rehabilitation Engineering Technology Research Center Construction Project of Guangdong Provincial Department of Education (2012gczxA1004).

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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