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Treatment of polluted river water by a gravel contact oxidation system constructed under riverbed

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ABSTRACT: The objective of this study was to evaluate the treatment efficiency of a gravel contact oxidation treatment system which was newly constructed under the riverbed of Nan-men Stream located at the Shin Chu City of Taiwan. The influent and effluent water samples were taken periodically for the analyses of pH, temperature, dissolved oxygen, total suspended solids, five-day biological oxygen demand, NH_4^+ -N. The results showed that the average removal rates of five-day biological oxygen demand, total suspended solids and NH_4^+ -N were 33.6% (between -6.7% and 82.1%), 56.3% (between -83.0% and 93.4%) and 10.7% (between -13.0% and 83.3%), respectively. The calculated mean first order reaction rate constant for five-day biological oxygen demand was 4.58/day with a standard deviation of 4.07/day and for NH_4^+ -N was 2.15/day with a standard deviation of 5.68/day. Therefore, it could be said that this gravel-contact-oxidation system could effectively remove biological oxygen demand, total suspended solids, and NH_4^+ -N in river water at a relatively short hydraulic retention time, although its pollutant treatment efficiency was not quite stable. However, to reach better or more stable treatment efficiency, aeration might sometimes be necessary to increase the dissolved oxygen in influent river water. And, longer hydraulic retention time of the system might also be required to increase NH_4^+ -N removal efficiency.

Key words: *Ecological engineering, gravel contact oxidation treatment system, loading rate, packed-bed reactor, water quality*

INTRODUCTION

A gravel contact oxidation treatment system is a kind of packed-bed reactor with the packed medium of gravels as biofilm carriers. It might be classified as one type of natural and ecological treatment techniques for the imropvement of river water quality. When the system is applied to treat the polluted river water, two installation ways are always seen (Crite *et al.*, 2000; Reed, 2000; Kivaisi, 2001; Zhen, 2002; Tsai, 2007). First one is installing the treatment system beside the river, and the other one is installing it inside the river. For the first way, the river water should be pumped or directed by gravity to the gravel contact oxidation treatment system located beside the river. However, for the second way, the river water normally flows by gravity through the gravel-packed-bed reactor. No matter

which way is selected, biofilm will grow on the surface of gravels and utilize the organic pollutants in the river water. Some researchers reported that the biofilm growing on the gravels will be thicker for an open channel with lower flow velocity (Lau, 1990; Lau and Liu, 1993).

The first order reaction equation shown as below could be used to express the removal of five-day biological oxygen demand (BOD₅) and NH₄⁺-N in a gravel-packed-bed reactor (Tanner, 1994; Reed *et al.*, 1995; USEPA, 2000; Dahab *et al.*, 2001; Dahab and Surampalli, 2001; Luederitz et al., 2001; Vymazal, 2002; Liu *et al.*, 2005):

$$\frac{C_e}{C_0} = e^{-K_t \times t} \tag{1}$$

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where C_0 (mg/L) and C_e (mg/L) are the pollutant concentrations in the influent and effluent, respectively. K_i /day is the first order reaction rate constant of pollutant and t(day) is the hydraulic retention time (HRT). This equation can also be expressed as the following one:

$$\ln(\frac{C_e}{C_0}) = -K_t \times t \tag{2}$$

Some researchers reported that the first order reaction rate constant varies significantly with the water velocities instead of being a constant as previously believed (Leu et al., 1996; Leu et al., 1998). In gravel packed-bed constructed wetlands, biochemical oxygen demand (BOD) and total suspended solids (TSS) removal could be very effective at a relatively short hydraulic retention time (HRT) and BOD removal exhibited a linear relationship with organic loading. Effective nitrogen removal required a longer HRT and appeared to be limited by the low oxygen availability in gravel packed-bed systems (Reed and Brown, 1995; Bergen et al., 2001; Coveney et al., 2002). The average removal rate of BOD was reported between 50 % and 70% for gravel contact oxidation treatment systems. Without aeration, the average BOD removal rate was normally in the range of 20 % - 70 %, and it was between 50 % and 80 % with aeration (Cooper and Findlater, 1990; Varrier and Dahab, 2001; Zhen, 2002; Fan and Wang, 2006). A study conducted by Hamersley et al. (2001) showed that the nitrogen removal in a gravel packed-bed constructed wetland was higher than 50% and was primarily by sedimentation of waste solids.

Yu et al. (2006) studied the treatment efficiency of a gravel contact oxidation treatment system located in Guandu, Taiwan. This system was constructed at the riverside. The river water was inducted into an influent well by piping, and then pumped to a storage tower by submersible pumps. Finally, the river water flew into the system by gravity. They reported that the BOD removal rate was ranged between 5.2 % and 79 % with an average of 46 %, the TSS removal rate was in a range between -134 % and 95.9 % with an average of 71%, and the NH_4^+ -N removal rate was ranged between -16.7 % and 59.1 % with an average of 24 %. They also obtained the K values for BOD and NH_4^+ -N were 1.4231/ day and 0.6132/day, respectively. These values were higher than those (= 0.3/day for BOD and 0.14/day for $NH_{4}^{+}-N$) obtained by other researchers in Europe (Luederitz et al., 2001). Normally, BOD removal rate

should be higher with the longer hydraulic retention time, however it should become stable with hydraulic retention time over 2 h. (Fan and Wang, 2006). However, Kadlec and Knight (1996) depicted that BOD removal rate higher than 70 % could only be obtained at the hydraulic retention time over 1.7days for gravel packedbed constructed wetlands. Meanwhiles, the size and the porosity of gravels were normally between 20 mm and 200 mm and between 30 % and 40 %, respectively (Reed et al., 1995; Kadlec and Knight, 1996; Spieles and Mitsch, 1999; Fan and Wang, 2006; Yu et al., 2006). Since Taiwan's Environmental Protection Administration (TWEPA) has been actively propagating the natural and ecological treatment techniques for the purification of river water, a new-built gravel contact oxidation treatment system was selected for study. In this study, a completed gravel contact oxidation treatment system under the river bed of a stream in the north of Taiwan was applied for the evaluation of water quality treatment efficiency. Since this gravel-packed-bed reactor could be claimed as the first one constructed inside the river and under the riverbed in Taiwan, many operational data and control criteria needed to be established. It is expected that the results obtained in this study could provide the operators with basic control criteria.

This research field of gravel contact oxidation treatment system was located at the Nan-men Stream in Shin Chu City, Taiwan. The system was constructed at the downstream and under the riverbed of this river and was completed in early November, 2006. This research was then conducted in situ starting from November, 2006 to May, 2007.

MATERIALS AND METHODS

Description of the gravel contact oxidation treatment system

The whole treatment system included a compound section of inlet channel, two bar screens, one grit chamber, three influent distribution channels, three effluent collection channels, and three gravel-packed contact oxidation tanks with the backwash air pipes and sludge collection channel installed at their bottoms. The whole system was constructed under the riverbed of Nan-men Stream located at the Shin Chu City, Taiwan. The design flow rate of this system was 10,000 CMD (m³/day), and it flew through the whole system by gravity. During clear days, the polluted river water will flow through inlet channel, pass through

two bar screens, then enter the grit chamber. At the end of grit chamber, three distribution weirs and three distribution channels are used to evenly distribute river water into three gravel-packed contact oxidation tanks. The treated water of each contact oxidation tank will flow through a collection channel and then back to the downstream of the river. During wet days, if the river flow rate is higher than the design flow rate, the superfluous flow will directly pass through the treatment system to the downstream of the river.

The volume of grit chamber is about 237.85 m³. The volume of each gravel-packed contact oxidation tank is about 434 m³ with the length of 31m, the width of 8 m, and the depth in a range of between 1.6 m and 1.9 m. Three contact oxidation tanks were operated in parallel. The gravels packed in the contact oxidation tanks had an average diameter between 50 mm and 150 mm, and had an average specific surface area of about 8 m²/m³. The average porosity among gravels after packed in contact oxidation tanks was about 43%. Therefore, the effective capacity of each contact oxidation tank was about 186.6 m³.

Analyses of water samples

After the gravel contact oxidation treatment system was constructed and stabilized for a few months, the influent and effluent water samples were collected and analyzed during a five-month period of time. The influent grab samples were taken at a location before distribution weirs. The effluent samples with the same volume were taken every time at the outlet of three collection channels and then mixed together as a compound sample for analysis. Due to the limitation of financial budget, water samples were only measured for water temperature, pH, dissolved oxygen (DO), BOD₅, TSS, and NH₄⁺-N, following the methods mentioned in Standard Methods (Clesceri et al., 2001). DO was measured on site by a DO meter (Hach DO meter - Model sensION6). BOD was determined by method 5210B of Standard Methods (HILES Incubator - Model LE-747), TSS was tested following method 2540 D of Standard Methods (MEMMERT Oven -Model ULM500), and NH₄-N was measured by an ammonium selective electrode following the procedure mentioned in method 4500 (Phenate Method) of Standard Methods (UNICO Spectrophotometer -Model SQ2800). For the confirmation of experimental accuracy, duplication of experimental analysis was applied to each water sample and the data from

duplicated tests of each water sample were then averaged.

Data analyses

The removal efficiencies (r, %) of pollutants were calculated as:

The mass loading rate (M_e , g/m²/day) was expressed as:

$$M_l = \frac{Co \times Q}{A}$$

where Co (mg/L) is the influent pollutant concentration, Ce (mg/L) is the effluent pollutant concentration, Q_i (m³/ day) is the influent flow rate, and A (m²) is the effective surface area of each treatment tank. In this study, all statistical analyses of the data were completed by using Excel or SPSS software (Juang and Chen, 2007) and the significance level of 0.05 was used in the ANOVA (Analysis of variance), the correlation, and the linear regression tests.

RESULTS AND DISCUSSION

The characteristics and water quality data of this treatment system were shown in Table 1. During the research period, the water temperature ranged between 15.4 °C and 27.2 °C in the influent and between 16.0 °C and 26.9 °C in the effluent. No obvious difference on pH values was seen between influents and effluents. Part of dissolved oxygen was consumed during treatment with an average DO consumption rate of 34% and a standard deviation (SD) of 26%.

Treatment efficiency

Figs. 1-3 showed the influent and effluent concentrations and the removal rates of BOD₅, TSS and NH₄⁺-N, respectively. The average removal rates of BOD₅, TSS, and NH₄⁺-N were 33.6% (between -6.7% and 82.1%) with a SD of 24.6%, 56.3% (between -83.0% and 93.4%) with a SD of 38.4%, and 10.69% (between -13.0% and 83.3%) with a SD of 33.4%, respectively. According to the Eqs. 1 or 2, the calculated mean first order reaction rate constant (K_t) for BOD₅ was 4.58/ day with a SD of 5.68/day. The values for both BOD and NH₄⁺-N in this gravel contact oxidation treatment system were much higher than those reported by Luederitz *et al.* (2001) and Yu *et al.* (2006).

Treatment efficiency of gravel contact oxidation system on river water

Water	Water quality Flow rate		Effective capacity of each	Ave. specific surface area of gravels	Effective	Temp (°C)		pH		DO (mg/L)	
(Items)	(CMD)	(h.)	unit (m ³)	(m^2/m^3)	surface (m ²) -	Inf.	Eff.	Inf.	Eff.	Inf.	Eff.
1	5320.0	2.5	186.6	8.0	1979.0	22.2	21.2	7.8	7.7	6.9	4.9
2	5602.0	2.4	186.6	8.0	1979.0	25.9	22.5	6.5	7.6	3.9	1.8
3	4880.0	2.8	186.6	8.0	1979.0	27.2	26.9	7.6	7.4	2.9	3.0
4	4702.0	2.9	186.6	8.0	1979.0	25.5	25.4	6.9	7.0	5.7	1.2
5	4563.0	2.9	186.6	8.0	1979.0	23.2	24.0	7.4	7.3	6.7	5.6
6	5330.0	2.5	186.6	8.0	1979.0	22.7	20.8	7.7	7.5	3.4	2.6
7	5607.0	2.4	186.6	8.0	1979.0	24.7	21.8	7.7	7.4	4.5	4.1
8	5433.0	2.5	186.6	8.0	1979.0	19.1	19.6	7.6	7.3	3.0	2.9
9	5225.0	2.6	186.6	8.0	1979.0	19.4	19.9	7.5	7.6	5.2	2.8
10	7556.0	1.8	186.6	8.0	1979.0	21.0	21.8	7.6	7.3	4.5	3.5
11	4333.0	3.1	186.6	8.0	1979.0	15.8	16.2	6.9	7.2	3.9	1.1
12	5232.0	2.6	186.6	8.0	1979.0	15.4	16.0	6.6	6.7	4.8	4.1
13	5055.0	2.7	186.6	8.0	1979.0	20.5	20.9	6.6	6.8	3.9	1.4
14	5218.0	2.6	186.6	8.0	1979.0	21.0	21.5	7.6	7.9	8.0	4.9
15	6531.0	2.1	186.6	8.0	1979.0	21.2	21.6	7.5	7.4	-	-
16	4425.0	3.0	186.6	8.0	1979.0	22.4	22.9	7.7	8.0	-	-
17	4826.0	2.8	186.6	8.0	1979.0	22.1	22.5	8.0	7.4	-	-
18	5305.0	2.5	186.6	8.0	1979.0	21.8	22.3	7.5	7.3	-	-
19	5863.0	2.3	186.6	8.0	1979.0	24.6	23.6	8.4	7.5	-	-
20	5001.0	27	186.6	8.0	1979.0	25.8	24.1	79	74	_	-

Table 1: Characteristics and water quality data of gravel contact oxidation system



Fig. 1: The influent and effluent concentrations and the removal rates of BOD_5

Fig. 2: The influent and effluent concentrations and the removal rates of TSS



Fig. 3: The influent and effluent concentrations and the removal rates of NH_4^+ -N

Relationship between pollutant loading rate and effluent concentration

Fig. 4 expressed a linear relationship between the effluent BOD concentration and the BOD mass loading rate of each gravel-contact-oxidation treatment tank. Although the result showed higher effluent BOD concentration with higher BOD mass loading, the coefficient of determination (R^2) was only 0.3876. This linear proportional relationship could be expressed as below:

$$Y = 0.4726 \times X + 2.7939 \tag{4}$$

where Y: effluent BOD concentration (mg/L) and X: BOD mass loading $(g/m^2/day)$.

Similarly, Figs. 5 and 6 also showed that a linear proportional relationship between the effluent TSS or

 NH_4^+ -N concentration and the TSS or NH_4^+ -N mass loading rate of each gravel-contact-oxidation treatment tank, respectively. The result also showed higher effluent TSS or NH_4^+ -N concentration with higher TSS or NH_4^+ -N mass loading, the coefficient of determination (R²) was 0.0163 for TSS and 0.5628 for NH_4^+ -N. Both linear relationships could be expressed as bellows:

$$Y = 0.1337 \times X + 6.2831 \tag{5}$$

where Y: effluent TSS concentration (mg/L) and X: TSS mass loading rate ($g/m^2/day$).

$$Y = 0.7678 \times X + 1.0047 \tag{6}$$

where Y: effluent NH_4^+ -N concentration (mg/L) and X: NH_4^+ -N mass loading rate (g/m²/day).



Fig. 4: Linear relationship between effluent BOD₅ concentration and BOD₅ mass loading rate of each gravel contact oxidation treatment tank



Fig. 5: Linear relationship between effluent TSS concentration and TSS mass loading rate of each gravel contact oxidation treatment tank



Fig. 6: Linear relationship between effluent NH_4^+-N concentration and NH_4^+-N mass loading rate of each gravel contactoxidation treatment tank



Fig. 8: Relationship between TSS removal rate and TSS mass loading rate of each gravel contact oxidation treatment tank

Relationship between pollutant loading rate and removal rate

Figs. 7-9 showed the relationships between BOD removal rate and BOD mass loading rate, between TSS removal rate and TSS mass loading rate, and between NH₄⁺-N removal rate and NH₄⁺-N mass loading rate, respectively, however their coefficients of determination (R²) were very low (R² = 0.1108 for BOD, R² = 0.0701 for TSS, and R² = 0.0097 for NH₄⁺-N). These linear equations could be expressed as bellows:

$$Y = 1.324 \times X + 16.565 \tag{7}$$

where Y: BOD_5 removal rate (%) and X: BOD mass loading rate (g/m²/day)



Fig. 7: Relationship between BOD₅ removal rate and BOD₅ mass loading rate of each gravel contact oxidation treatment tank



Fig. 9: Relationship between NH₄⁺-N removal rate and NH₄⁺-N mass loading rate of each gravel contact oxidation treatment tank

$$Y = 1.3713 \times X + 30.125 \tag{8}$$

where Y: TSS removal rate (%) and X: TSS mass loading rate $(g/m^2/day)$

$$Y = 0.9771 \times X + 6.0272 \tag{9}$$

(0)

where Y: NH_4^+ -N removal rate (%) and X: NH_4^+ -N mass loading rate ($g/m^2/day$).

Relationship between HRT and pollutant removal rate or effluent concentration

Fig. 10 expressed a relationship between hydraulic retention time (HRT) and pollutant removal rates of each treatment unit and apparently no obvious

differences on the removal rates of BOD, TSS and NH_4^+ -N were seen in the range of HRT (from 1.8 h. to 3.1 h.). Fig. 11 illustrated the relationship between hydraulic retention time (HRT) and effluent pollutant concentration of each treatment unit. Similarly, no good relationship between them was concluded. Kadlec and Knight (1996) mentioned that BOD removal rate higher than 70 % could only be obtained with the hydraulic retention time over 1.7 days. Kemp and George (1997) also reported that the effluent NH⁺₄-N concentration could have obvious reduction when the HRT of subsurface flow constructed wetland was increased from 1.7 days to 3.9 days. However, Reed and Brown (1995) claimed that BOD removal could be very effective at a relatively short HRT and effective nitrogen removal might require a longer HRT. Although this treatment system showed certain degree of treatment efficiency on pollutants at lower HRTs, further studies might be

required to confirm whether higher HRT will improve the treatment efficiency of gravel contact oxidation system. The ANOVA test results in Table 2 showed that there were no significant differences (p > 0.05) on the removal rates of BOD₅ and NH₄⁺-N with the HRT less than 2.5 h., between 2.5 h. and 2.8 h. and higher than 2.8 h. However, significant differences (p = 0.027) on the removal rates of TSS were seen with the HRT less than 2.5 h., between 2.5 h. and 2.8 h. and higher than 2.8 h. The correlations between the effluent concentrations or the removal rates of pollutants and the HRT or the mass loading rates of gravel-packed reactors were shown in Table 3. Apparently, effluent BOD concentration and its mass loading rate had a significant correlation (p = 0.003) in the treatment system. Similar result was seen between effluent NH⁺-N concentration and its mass loading rate with the p value of about 0.003.



Fig. 10: Relationship between pollutant removal rate and hydraulic retention time



Fig. 11: Relationship between effluent pollutant concentration and hydraulic retention time

Parameter	HRT (h.)	n	Average removal rate (%)	SD (%)	F	P-value
BOD ₅	≤2.5	9	25.4	23.7	0.909	0.422
	>2.5 and <2.8	5	39.5	19.9		
	≥ 2.8	6	40.9	29.4		
	Total	20	33.6	24.6		
TSS	≤2.5	9	70.6	17.2	4.518	0.027
	>2.5 and <2.8	5	18.2	59.5		
	≥ 2.8	6	66.8	19.0		
	Total	20	56.3	38.4		
NH4 ⁺ -N	≤2.5	6	14.8	33.6	0.850	0.456
	>2.5 and <2.8	3	-11.1	13.6		
	≥ 2.8	4	20.9	42.5		
	Total	13	10.7	33.4		

Table 2: ANOVA tests for pollutant removal rates at different hydraulic retention time

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Table 3: Correlations between effluent pollutant concentrations or removal rates and HRTs or pollutant loading rates

Treatment characteristics	HRT	Pollutant loading rate		
Eff. BOD concentration	0 310 (-0 239)	0.003 (0.623)		
P-value (pearson correlation coefficient)	0.510 (0.255)	0.005 (0.025)		
Eff. TSS concentration	0 135 (0 346)	0 593 (0 127)		
P-value (pearson correlation coefficient)	0.155 (0.540)	0.393 (0.127)		
Eff. NH_4^+ -N concentration	0.802(0.042)	0.002 (0.752)		
P-value (pearson correlation coefficient)	0.893 (-0.042)	0.003 (0.732)		
BOD removal rate	0.544 (0.144)	0 152 (0 222)		
P-value (pearson correlation coefficient)	0.344 (0.144)	0.155 (0.552)		
TSS removal rate	0.666 (0.102)	0.261 (0.264)		
P-value (pearson correlation coefficient)	0.000 (-0.103)	0.201 (0.204)		
Eff. NH_4^+ -N removal rate	0.530 (0.102)	0.750 (0.000)		
P-value (pearson correlation coefficient)	0.530 (0.192)	0.750 (0.098)		



concentration and DO consumption

Relationship between DO consumption and pollutant removal rate or effluent concentration

Figs. 12 and 13 illustrated the relationships between effluent pollutant concentration and DO consumption and between pollutant removal rate and DO consumption in each treatment unit. The result showed that only NH_4^+ -N removal rate had a better linear relationship with DO consumption in the gravelcontact-oxidation treatment tank, with the coefficient of determination (R²) of 0.2431, and the linear relationship could be expressed as below:

$$Y = 12.561 \times X - 9.3184 \tag{10}$$

where Y: NH_4^+ -N removal rate (%) and X: DO consumption (mg/L)

This gravel contact oxidation treatment system was the first one constructed under riverbed in Taiwan.



Fig. 13: Relationship between pollutant removal rate and DO consumption

Since the river water flew through this system by gravity, no power was consumed in the whole treatment process and the operation and maintenance cost was apparently reduced. However, DO in the influent seemed to be unstable and this might cause a labile treatment efficiency in the system. According to the water quality results, the removal rates of BOD₅, TSS and NH⁺-N varied significantly. With the HRT range (1.8-3.1 h.) applied to each treatment unit in this study, it is difficult to make good conclusions on the relationship between pollutant removal rates or effluent pollutant concentration and HRT. Since this research started a few weeks after this treatment system was completely constructured, it is possible that the biofilm growing on the gravels has not yet developed well and the treatment system has not yet reached fully stable during the research period. Therefore, further studies will be required to obtain more water quality data and compare the results found in this study.

Basically, the average removal rates of BOD, TSS and NH_4^+ -N were 33.6 % (between -6.7% and 82.1%), 56.3 % (between -83.0% and 93.4 %) and 10.7% (between -13.0 % and 83.3 %), respectively. The BOD₅ removal rates found in this gravel contact oxidation treatment system without aeration seemed to be reasonable according to the range of 20% - 70% described by Fan and Wang (2006). However, the average BOD and NH_4^+ -N removal rates (33.6 % and 10.7 %, respectively) in this system were somewhat lower than those (BOD = 46% and 24%, respectively) reported by Yu et al. (2006). In this gravel contact oxidation treatment system, the effluent BOD concentration should be higher if the BOD mass loading rate was higher. This means that a linear proportional relationship was found between effluent BOD concentration and BOD mass loading rate ($R^2 = 0.3876$). Similarly, a linear proportional relationship was also found between effluent NH⁺-N concentration and NH₄⁺ N mass loading rate $(R^2=0.5628)$. Basically, the linear relationships between BOD and NH⁺₄-N removal and their loading rates were coincident to the conclusion reported by other researchers (Reed and Brown, 1995; Dahab and Surampalli, 2001; Varrier and Dahab, 2001). By the way, no significant differences (p > 0.05) were seen on the removal rates of BOD₅ and NH₄⁺-N with the HRT less than 2.5 h., between 2.5 h. and 2.8 h. and higher than 2.8 h. However, significant differences (p < 0.05) were concluded on the TSS removal rates with HRT less than 2.5 h., between 2.5 h. and 2.8 h. and higher than 2.8 h. Effluent BOD or NH₄-N concentration had a significant correlation (p = 0.003) with its mass loading rate in the treatment system. A significant correlation was also seen between TSS removal rate and its mass loading rate (p = 0.003). It is also found that NH_4^+ -N removal rate had a better linear proportional relationship with DO consumption in the gravelcontact-oxidation treatment tank, with the coefficient of determination (R^2) of 0.2431.

According to the results and discussion abovementioned, this gravel contact oxidation treatment system should be able to effectively remove BOD, TSS and NH_4^+ -N in river water at a relatively short HRT, although its pollutant treatment efficiency was not quite stable. The dissolved oxygen aeration might sometimes be required to increase the dissolved oxygen in influent river water and remain a stable treatment efficiency of pollutants in the system. By the way, further studies might be required to confirm whether higher HRT will improve the treatment efficiency of this gravel contact oxidation system.

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D. F. Juang et al.

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