


Provided by the author(s) and University College Dublin Library in accordance with publisher policies. Please cite the published version when available.

Title	Enhanced removal of organic matter and ammoniacal-nitrogen in a column experiment of tidal flow constructed wetland system
Author(s)	Sun, Guangzhi; Zhao, Y.Q.; Allen, Stephen
Publication date	2005-01-26
Publication information	Journal of Biotechnology, 115 (2): 189-197
Publisher	Elsevier
Link to online version	http://dx.doi.org/10.1016/j.jbiotec.2004.08.009
Item record/more information	http://hdl.handle.net/10197/3172
Publisher's statement	This is the author's version of a work that was accepted for publication in Journal of Biotechnology. Changes resulting from the publishing process, such as peer review, editing, corrections, structural formatting, and other quality control mechanisms may not be reflected in this document. Changes may have been made to this work since it was submitted for publication. A definitive version was subsequently published in Journal of Biotechnology, 115 : 189-197 DOI 10.1016/j.jbiotec.2004.08.009.
Publisher's version (DOI)	http://dx.doi.org/10.1016/j.jbiotec.2004.08.009

Downloaded 2018-03-23T16:59:26Z

The UCD community has made this article openly available. Please share how this access benefits you. Your story matters! (@ucd_oa) 

Some rights reserved. For more information, please see the item record link above.



Enhanced removal of organic matter and ammoniacal-nitrogen in a column experiment of tidal flow constructed wetland system

Guangzhi Sun^{*}, Yaqian Zhao, Stephen Allen

School of Chemical Engineering, Queen's University Belfast, David Keir Building, Stranmillis Road, Belfast BT9 5AG, Northern Ireland, UK

^{*} Correspondence. Tel: +44-(0)28-90274692; Fax: +44-(0)28-90381753; E-mail: G.Sun@qub.ac.uk

Abstract

This study investigated the efficiency of a four-stage tidal flow constructed wetland system for the removal of organic matter and ammoniacal-nitrogen from diluted piggery wastewater. The results demonstrated that the operation of tidal flow enhanced the transfer of oxygen into wetland matrices. The overall oxygen consumption rate in the tidal flow system ($357 \text{ gO}_2/\text{m}^2\cdot\text{d}$) was considerably higher than the rate obtainable in conventional wetlands. Most oxygen consumption (99%) was due to the decomposition of organic matter. The total supply of oxygen into the wetlands ($473 \text{ gO}_2/\text{m}^2\cdot\text{d}$) exceeded the demand for the treatment. The percentage removal of BOD_5 and $\text{NH}_4\text{-N}$ was improved by effluent recirculation at a ratio of 1:1. Immobilization by microbial cells and adsorption were found to be the main routes for the removal of ammoniacal-nitrogen. Significant nitrification could not take place under the treatment condition of the experiment.

Keywords: Ammonia; Biofilm; BOD_5 ; Reed bed; Wastewater treatment

1. Introduction

The efficiency of pollutant removal varies considerably in different constructed wetlands used for the treatment of wastewaters. After nearly two decades of gradual improvement, the removal of organic substances (typically 80-99%) is now satisfactory in most treatment wetlands. However, the removal of inorganic nitrogen is often unsatisfactory (Luederitz et al., 2001). In Europe, typical percentage removal of ammoniacal-nitrogen during long-term operation is only 35% or, up to 50%, after some modifications are made to stimulate the transformation of nitrogen (Verhoeven and Meuleman, 1999). In particular, the removal of ammoniacal-nitrogen from strong wastewaters appears to be inadequate in most wetlands.

Several studies have shown improvement in the performance of treatment wetlands by adopting an innovative operation known as “tidal flow” or “reciprocation” (Green et al., 1997; Sun et al., 1999;

Leonard et al., 2003). During the tidal flow operation, the matrix of wetland is alternately filled and drained with wastewater. As the wetland is filled, air is repelled from the matrix as the wastewater level rises. When the wetland is drained, the retreating wastewater acts as a passive pump to draw air from the atmosphere into the matrix, thereby creating a cycle of “wet” and “dry” period in the wetland. Tidal flow operation has shown the potential to enhance the removal of BOD₅ through aerobic decomposition and the removal of ammoniacal-nitrogen through nitrification/denitrification, because maximum pollutant-biofilm contact is established and oxygen transfer rate is increased during the operation (Sun et al., 1999; Leonard et al., 2003).

Another challenge facing researchers in constructed wetland is to extend its application to strong effluents, e.g. dairy farm wastewater, landfill leachate and effluent from slaughter house. The presence of a large amount of carbonaceous substrates in these strong effluents often makes oxygen unavailable for nitrification in the wetland. Inadequate pollutants-microorganisms contact can also inhibits nitrification because nitrifying bacteria are autotrophs that have a slower respiration rate than the heterotrophs which consume organic substrates (Sikora et al., 1995; Reddy and D’Angelo, 1997). It is necessary to adopt vertical flow pattern and force the effluents to recirculate around the wetland in order to facilitate the treatment of strong effluents. In literature, Sikora et al. (1995), Kantawanichkul et al. (2001), Green et al. (2002), Brix et al. (2002) and Sun et al. (2003) all reported the beneficial effect of effluent recirculation on the performance of wetlands, especially on the removal of inorganic nitrogen. Kantawanichkul and Neamkam (2001) studied the variation of recirculation ratio (recirculation flow rate:feed flow rate) from 0:1 to 2:1 in a vertical flow wetland system treating pig farm wastewater. The optimum ratio was found to be 1:1 which allowed the highest nitrogen removal, 93% Total Nitrogen, to be achieved in the system. Through a lab-scale trial, this study aims to investigate the performance of a tidal flow wetland system treating strong effluent and, more importantly, discover the mechanism of nitrogen removal and the effect of recirculation on the treatment process.

2. Materials and methods

The wetland system consisted of four stages, each being operated with the tidal flow strategy. Each stage consisted of a single lab-scale wetland that was constructed with Perspex columns of 95 mm in diameter and 900 mm in height. The columns were filled with 26.4 ± 7.2 mm round gravel to a depth of 150 mm as bottom layer and main filter layer of 4.4 ± 1.5 mm gravel with a depth of 650 mm. A single young *Phragmites australis* from a local commercial supplier was planted in the main layer of each wetland, as shown in Fig. 1.

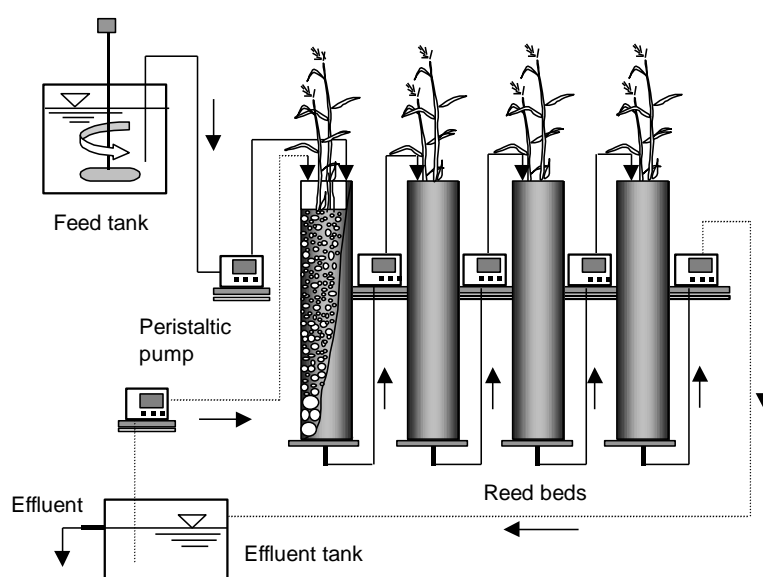


Fig. 1 Schematic description of the constructed wetland system

Wastewater was prepared in a feed tank by diluting raw pig slurry collected from a local farm. After collection, the raw slurry was allowed to settle overnight and the supernatant with initial BOD_5 of 37.3-92.4 g/l was diluted with tap water. From the feed tank diluted slurry was batch loaded into the first stage and passed through the system sequentially from the first to the fourth stages. The 'tide', cycles of wet/dry period, was generated in each stage by peristaltic pumps controlled by a timer. Based on results from a previous study (Zhao et al., 2003) the cycle was set to take place every four hours, giving each wetland one hour of wastewater-media contact and three hours of aeration period in each cycle.

Experiments were conducted in two periods. In the first period, which lasted for seven weeks, the wastewater passed through the wetland system without any recycling. This was followed by the second period that lasted for approximately nine weeks when effluent recirculation was employed at a recirculation ratio of 1:1. Hydraulic loading onto the overall system was set at $0.43 \text{ m}^3/\text{m}^2 \cdot \text{d}$ (m^2 represents the total surface area of the system) throughout the experiment. The wetlands were left resting for one week after each week of operation in both periods.

Wastewater samples were collected from each stage when the system was operating. The samples were analysed for BOD_5 , $\text{NH}_4\text{-N}$, $\text{NO}_2\text{-N}$, $\text{NO}_3\text{-N}$ and pH. A Piccolo II portable pH meter was used for the pH analysis. BOD_5 was determined using a BODTrack apparatus (CAMLAB Ltd., UK). $\text{NH}_4\text{-N}$ was tested using a Sension II pH/ISE meter and an ammonia electrode. The remaining parameters were analysed with a HACH DR2010 Colorimeter (CAMLAB Ltd, UK) according to its standard operation procedure.

3. Results

A total of twelve sets of data were collected during the first period of the experiment. Another ten sets were collected during the second period. Fig. 2 illustrates the changes of BOD_5 , ammoniacal-, nitrite- and nitrate-nitrogen values across the whole wetland system in these periods. In order to highlight the changes of these values, all parameters in Fig. 2 are presented as ratios of inlet concentrations into the first stage (C_0) over the outlet concentrations from the final stage (C_e). Average percentage removals of BOD_5 and $\text{NH}_4\text{-N}$ are presented in Fig. 3. Table 1 shows the mean treatment results in each stage of the wetland system.

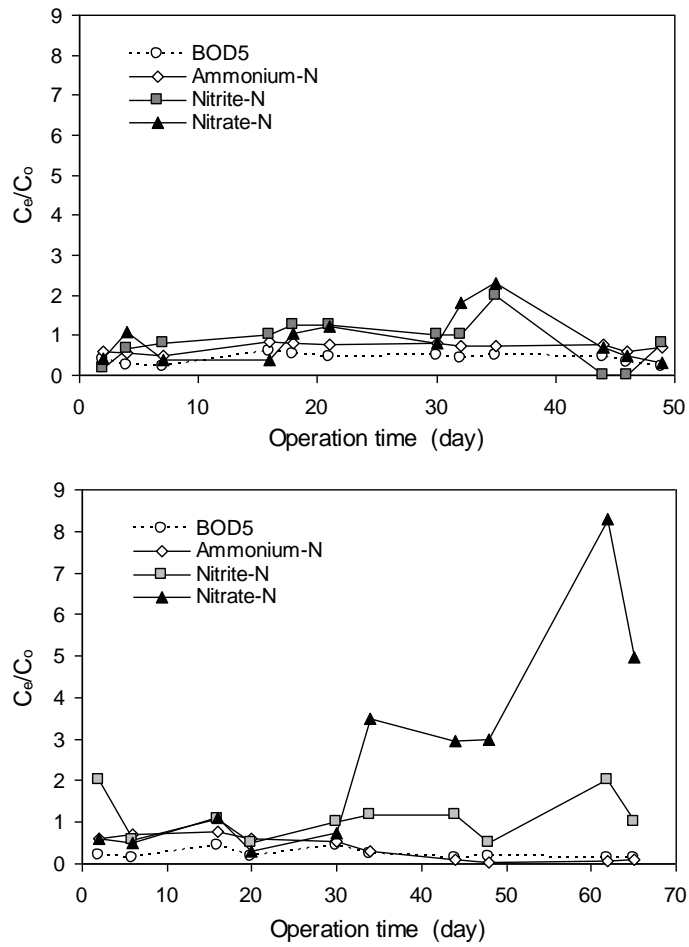


Fig. 2 Variation of BOD₅ and nitrogen levels in Period 1 (upper) and Period 2 (lower)

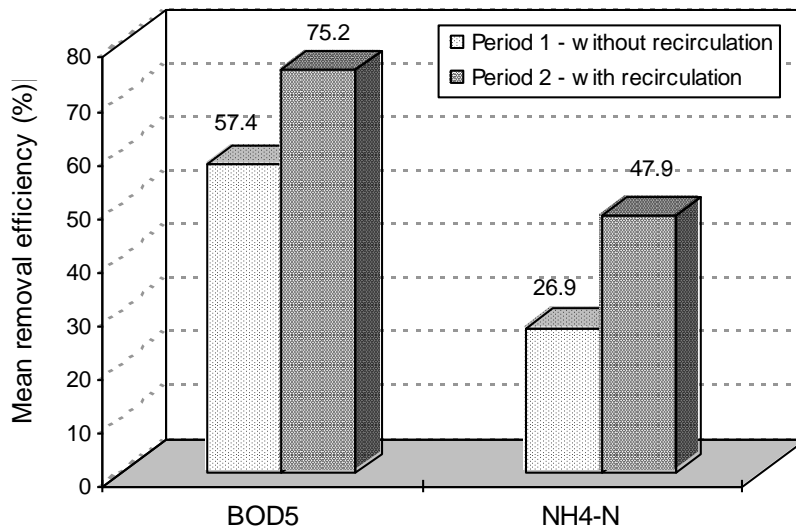


Fig. 3 Average percentage removal of BOD₅ and NH₄-N in the wetland system

Table 1 Mean concentrations (mg/l, excluding pH) in individual stages

Parameter	Influent	First stage	Second stage	Third stage	Fourth stage
Without effluent recirculation (N = 12)					
BOD ₅	2157	1716	1450	1142	918
NH ₄ -N	104	98	90	81	76
NO ₂ -N	2.2	2.1	3.2	3.0	2.1
NO ₃ -N	3.6	3.4	3.2	2.8	2.7
pH	6.9	7.0	7.2	7.3	7.5
With effluent recirculation (N = 10)					
BOD ₅	1359	1021	812	585	337
NH ₄ -N	121	102	95	77	63
NO ₂ -N	5.4	6.8	6.5	7.2	5.2
NO ₃ -N	3.4	4.2	6.0	8.9	9.0
pH	6.9	6.8	6.8	6.8	6.8

4. Discussion

4.1. The operation of tidal flow and effluent recirculation

Fig. 3 shows that the efficiency of the wetland system was improved in Period 2 as a result of effluent recirculation. The percentage removal of BOD₅ was increased by 17.8%, as demonstrated in Fig. 3. The bulk of ammoniacal-nitrogen was not nitrified in either period. However, in Period 2 there was sign of nitrification taking place as the levels of nitrite and nitrate in the wastewater started to rise, as indicated in Fig. 2. The reason for the improvement in ammoniacal-nitrogen removal in Period 2 may be attributed to prolonged wastewater-media contact in the wetland system resulted from the recirculation, as nitrifying bacteria are autotrophic microorganisms that multiply slowly and may need considerable time to convert ammonium into nitrite and nitrate (Sikora et al., 1995; Reddy and D'Angelo, 1997). Some other studies on municipal and agricultural wastewaters have also shown that effluent recirculation enhances nitrogen removal. Table 2 compares results from four different studies with regard to the effect of recirculation on the removal of ammoniacal- or total-nitrogen.

Table 2 The effect of recirculation on the removal of nitrogen (NH₄-N or TN)

Wetland systems	Wastewater	Recirculation ratio	Nitrogen removal efficiency (%)		Reference
			Without recirculation	With recirculation	
Two-stage vertical flow system	Municipal	1:1	48 (TN)	64 (TN)	Brix et al., 2002
Three-stage vertical flow system	Agricultural	20:1	19 (NH ₄ -N)	70 (NH ₄ -N)	Sun et al., 2003
Combined vertical and horizontal flow system	Pig farm	2:1	71 (TN)	85 (TN)	Kantawanichkul et al., 2001
Four-stage tidal flow system	Pig farm	1:1	27 (NH ₄ -N)	48 (NH ₄ -N)	This study

It should be noted that the hydraulic loading, 0.43 m³/m²·d, and organic loading, 594-915 gBOD₅/m²·d, on the wetland system was very high during the experiment. Typical loadings on most conventional wetlands are considerably lower. Brix (1994) recommended hydraulic loading range between 0.025-0.050 m³/m²·d for vertical flow wetlands. Weedon (2003) reported effectively operating a compact vertical flow wetland with BOD₅ loading up to 14 g/m²·d. Despite the high loading rate in this study, considerable amount of organic matter and ammoniacal-nitrogen was removed and the percentage removal of NH₄-N was reasonably comparable with the removal obtained from other wetland systems that were under lower loadings. The extensive removal of organic matter and ammoniacal-nitrogen achieved in this study is a result of the tidal flow operation that allowed maximum wastewater-media contact and enhanced the transfer of oxygen by drawing air from the atmosphere into the wetland matrices.

4.2. The route of NH₄-N removal

Several studies have reported that the reduction of ammoniacal-nitrogen is not balanced by increase in nitrite- and nitrate-nitrogen concentration in wastewater in the constructed wetlands (Tyrrel et al., 2002; Sun et al., 2003). Although the ‘disappearance’ of ammonia is likely to be a collective result of various processes including plant uptake and volatilization, it is believed that these two processes may only make a minor contribution to the removal of ammoniacal-nitrogen. The process of immobilization into microbial cells, i.e. biomass assimilation, has not received much attention with

regard to the removal of inorganic nutrients in wetlands. Because a large amount of organic matter was removed by the growth and respiration of microorganisms during the experiment, the process of biomass assimilation may have played a significant role in the removal of ammoniacal-nitrogen, considering that nitrogen constitutes a major part of the biomass, e.g. 12.4% of the mass of $C_5H_7O_2N$ being nitrogen (Cannon et al., 2000). It is therefore reasonable to conclude that nitrification, adsorption onto wetland media and immobilizations into microbial cells are the main routes of ammonia removal in the current wetland system. A conceptual model to describe the routes is illustrated in Fig. 4.

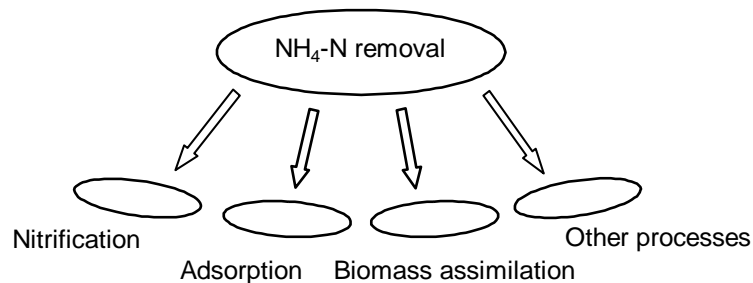


Fig. 4 The route of NH_4-N removal in the wetland system

It would be possible to quantify the contributions to the overall removal of ammoniacal-nitrogen by the processes shown in Fig. 4, provided that valid data are available for the calculation of adsorption isotherm and biomass assimilation. However, existing information on adsorption and assimilation processes is very limited and often contradicting, e.g. the adsorption capacity of ammonium onto gravel media was estimated as 175 mg NH_4-N/kg gravel by Sikora *et al* (1995), whereas the capacity was reported as 500 mg NH_4-N/kg gravel in another report (Stein et al., 2003). It is therefore difficult to precisely quantify the transformation of ammoniacal-nitrogen via the routes indicated in Fig. 4. Nevertheless, the contribution by biomass assimilation should be considered significant in this study, as Table 1 shows that a large amount of organic matter is removed from the wastewater in both periods of the experiment. The process of organic matter removal produces biomass. It has been estimated that 0.6 g biomass is generated as 1.0 g BOD_5 is removed (Cannon et al., 2000);

based on this figure and the approximation that nitrogen constitutes 12.4% of the biomass, the amount of ammoniacal-nitrogen immobilized by biomass assimilation is up to 0.074 g for each gram of BOD₅ removal. However, the actual figure should be lower than 0.074 g considering that ammonium is released into the wastewater by ammonification when the biomass is decomposed.

The increase in the concentration of nitrite and nitrate in the wastewater is very limited, as shown in Table 1. Therefore nitrification is not a major process to remove ammoniacal-nitrogen in this study. A mass balance calculation using concentrations of ammonium, nitrite and nitrate shows that less than 10% of NH₄-N removal was due to nitrification. The high content of organic matter in the wastewater may have inhibited nitrification, as oxygen available in the wetland is mostly used by heterotrophic microorganisms to remove organic matter and significant nitrification cannot take place until BOD₅ drops to 200 mg/l or below (Su and Ouyang, 1996; Sun et al., 1998).

4.3. Oxygen supply and consumption

The biological removal of BOD₅ and ammoniacal-nitrogen is oxygen-consuming. The supply of oxygen is therefore a critical issue concerning constructed wetlands, particularly in the treatment of strong wastewaters. In this study the operation of tidal flow is adopted to facilitate the transfer of oxygen. In the constructed wetlands oxygen can be provided and consumed via different routes. Fig. 5 highlights the main routes.

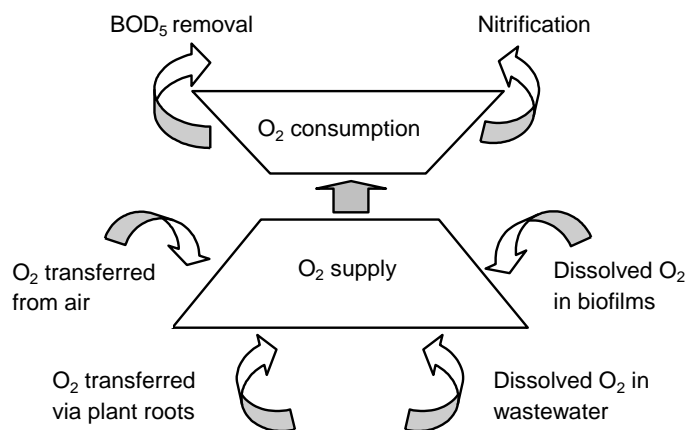


Fig. 5 The main routes of oxygen supply and consumption in constructed wetland

During the tidal flow operation, the volume of air supply depends on the pore space of wetland matrix. Accordingly, Green et al. (1997) claimed that as wastewater is drained the oxygen supply (mg) via air flow is $279.4 \times V$, where V represents the pore space (litre). Oxygen in biofilms growing on gravel surfaces can be estimated according to the following assumptions (Sun et al., 1998): the thickness of the biofilms being 1 mm with water content of 95%; the biofilms being saturated with dissolved oxygen (9.2 mg/l at 20°C); the specific surface area in the matrix being $100 \text{ m}^2/\text{m}^3$. Consequently, oxygen available in the biofilms amounts to $0.874 \text{ g}/\text{m}^3$ -matrix before the wastewater flowing into the wetlands. The amount of oxygen dissolved in strong effluents is usually negligible. Regarding the transfer of oxygen via plant roots, Brix (1990) suggested that oxygen released from the roots into the rhizosphere of common reeds is lower than $0.02 \text{ gO}_2/\text{m}^2 \cdot \text{d}$. Although this figure may be debatable, it can be ascertained that in tidal flow wetlands the amount of oxygen transfer via the plants is very small compared with the supply from the air flux. Regarding the consumption of oxygen, theoretically 1 g O_2 is required to remove 1 g BOD_5 , and up to 4.35 g O_2 to transform 1 g ammoniacal-nitrogen into nitrate. Table 3 presents an estimation of oxygen supply and consumption rate in the current wetland system during the whole experiment period.

Table 3 Oxygen supply and consumption in the wetland system

	O_2 consumption for BOD_5 reduction ($\text{g}/\text{m}^2 \cdot \text{d}$)	354
O_2 consumption	O_2 consumption for nitrification ($\text{g}/\text{m}^2 \cdot \text{d}$)	3
	Total O_2 consumption ($\text{g}/\text{m}^2 \cdot \text{d}$)	357
	O_2 supply from air flux ($\text{g}/\text{m}^2 \cdot \text{d}$)	469
O_2 supply	O_2 supply from biofilms ($\text{g}/\text{m}^2 \cdot \text{d}$)	4
	Total O_2 supply ($\text{g}/\text{m}^2 \cdot \text{d}$)	473

As demonstrated in Table 3, the supply of oxygen ($473 \text{ gO}_2/\text{m}^2 \cdot \text{d}$) from the air and the biofilms exceeds the demand ($357 \text{ gO}_2/\text{m}^2 \cdot \text{d}$). Most oxygen (99%) has been used for the removal of BOD_5 . The oxygen consumption rate is considerably higher than conventional horizontal flow and downflow wetlands, demonstrating the advantage of the tidal flow operation. The air drawn from

the atmosphere into the wetland matrices during the tidal flow operation is the predominant oxygen resource that accounts for over 99% of the supply. As a certain amount of oxygen can also be transferred into the wetland system via the roots of the reeds and by the wastewater, the total supply of oxygen is likely to be higher than $473 \text{ gO}_2/\text{m}^2\cdot\text{d}$. Nevertheless, the supply is sufficient to sustain the wastewater treatment in the tidal flow wetland system.

5. Conclusions

(1) Significant removal of organic matter and ammoniacal-nitrogen was achieved in a column trial of tidal flow constructed wetland system treating diluted piggery wastewater. The operation of effluent recirculation enhanced the treatment.

(2) The predominant processes for the removal of ammoniacal-nitrogen were immobilization by microbial cells and adsorption. Significant nitrification did not take place as the effluent from the wetland system still contained high level of organic matter.

(3) The oxygen consumption rate achieved in the system, $357 \text{ gO}_2/\text{m}^2\cdot\text{d}$, was considerably higher than the rate obtained in the wetlands operated with conventional methods. An estimation of oxygen supply and consumption showed that the supply to the system exceeded the demand. Most oxygen was transferred from the atmosphere into the wetlands by the tidal flow operation.

Acknowledgements

The authors acknowledge financial support from the Engineering and Physical Sciences Research Council (EPSRC) in the UK that allows this work to be carried out. Hall's Farm is thanked for supplying piggery wastewater.

References

Brix, H., 1990. Gas exchange through the soil-atmosphere interphase and through dead culms of *Phragmites australis* in a constructed reed bed receiving domestic sewage. *Water Res.* 24, 259-266.

Brix, H., 1994. Use of constructed wetland in water pollution control: historical development, present status, and future perspectives. *Water Sci. Technol.* 30, 209-223.

Brix, H., Arias, C.A., Johansen, N.H., 2002. BOD and nitrogen removal from municipal wastewater in an experimental two-stage vertical flow constructed wetland system with recycling. In: *Proceedings of the 8th International Conference on Wetland Systems for Water Pollution Control*. Dar es Salaam, Tanzania, pp. 400-410.

Cannon, A.D., Gray, K.R., Biddlestone, A.J., Thayanithy, K., 2000. Pilot-scale development of a bioreactor for the treatment of dairy dirty water. *J. Agric. Eng. Res.* 77, 327-334.

Green, M., Friedler, E., Ruskol, Y., Safrai, I., 1997. Investigation of alternative method for nitrification in constructed wetlands. *Water Sci. Technol.* 35, 63-70.

Green, M., Gidron, E., Lahav, O., Tarre, S., 2002. Treatment of dairy wastewater using a vertical bed with passive aeration. In: *Proceedings of the 8th International Conference on Wetland Systems for Water Pollution Control*. Dar es Salaam, Tanzania, pp. 324-332.

Kantawanichkul, S., Neamkam, P., 2001. Optimum recirculation ratio for nitrogen removal in a combined system: vertical flow vegetated bed over horizontal flow sand bed. In: *4th International Workshop on Nutrient Recycling and Retention in Natural and Constructed Wetlands*. Trebon, Czech Republic.

Kantawanichkul, S., Neamkam, P., Shutes, R.B.E., 2001. Nitrogen removal in a combined system: vertical flow vegetated bed over horizontal flow sand bed. *Water Sci. Technol.* 44, 137-142.

Leonard, K.M., Key, S.P., Srikanthan, R., 2003. A comparison of nitrification performance in gravity-flow and reciprocating constructed wetlands. In: Brebbia, C.A., Almorza, D. and Sales, D. (Eds.), *Water Pollution VII: Modelling, Measuring and Prediction*. WIT Press, Southampton, UK, pp. 293-301.

Luederitz, V., Eckert, E., Lange-Weber, M., Lange, A., Gersberg, R.M., 2001. Nutrient removal efficiency and resource economics of vertical flow and horizontal flow constructed wetlands. *Ecol. Eng.* 18, 157-171.

Reddy, K.R., D'Angelo, E.M., 1997. Biogeochemical indicators to evaluate pollutants removal efficiency in constructed wetlands. *Water Sci. Technol.* 35, 1-10.

Sikora, F.J., Tong, Z., Behrends, L.L., Steinberg, S.L., Coornod, H.S., 1995. Ammonium removal in constructed wetlands with recipulating subsurface flow: removal rates mechanisms. *Water Sci. Technol.* 32, 193-202.

Stein, O.R., Hook, P.B., Biederman, J.A., Allen, W.C., Borden, D.J., 2003. Does batch operation enhance oxidation in subsurface constructed wetlands? *Water Sci. Technol.* 48, 149-156.

Su, J.L., Ouyang, C.F., 1996. Nutrient removal using a combined process with activated sludge and fixed biofilm. *Water Sci. Technol.* 34, 477-486.

Sun, G., Gray, K.R., Biddlestone, A.J., 1998. Treatment of agricultural wastewater in downflow reed beds: experimental trials and mathematical model. *J. Agric. Eng. Res.* 69, 63-71.

Sun, G., Gray, K.R., Biddlestone, A.J., 1999. Treatment of agricultural wastewater in a pilot-scale tidal flow reed bed system. *Environ. Technol.* 20, 233-237.

Sun, G., Gray, K.R., Biddlestone, A.J., Allen, S.J., Cooper, D.J., 2003. Effect of effluent recirculation on the performance of a reed bed system treating agricultural wastewater. *Process Biochem.* 39, 351-357.

Tyrrel, S.F., Leeds-Harrison, P.B., Harrison, K.S., 2002. Removal of ammoniacal nitrogen from landfill leachate by irrigation onto vegetable treatment planes. *Water Res.* 36, 291-299.

Verhoeven, J.T.A., Meuleman, A.F.M., 1999. Wetlands for wastewater treatment: opportunities and limitations. *Ecol. Eng.* 12, 5-12.

Weedon, C.M., 2003. Compact vertical flow constructed wetland systems - first two year's performance. *Water Sci. Technol.* 48, 15-23.

Zhao, Y.Q., Sun, G., Lafferty, C., Allen, S.J., 2003. Optimising performance of a novel reed bed system for the treatment of high strength agricultural wastewater. In: *Asian Waterqual 2003*. Bangkok, Thailand.