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**Abstract**

The concept, design and performance analysis of a four-stage novel constructed wetland system (CWs) capable of enhanced and simultaneous removal of phosphorus (P) and organic matter (OM) from wastewaters is described. Alum sludge, a largely available by-product of drinking water facilities using aluminium salts as coagulant was used as the media. Under a hydraulic loading rate of 1.27 m$^3$/m$^2$.d and a range of organic loading rate of 279.4–774.7 g-BOD$_5$/m$^2$.d and 361.1–1028.7 g-COD/m$^2$.d, average removal efficiencies (mean±SD) of 90.6±7.5% for BOD$_5$ and 71.8±10.2% for COD were achieved, respectively. P removal was exceptional with average removal efficiency of 97.6±1.9% achieved for soluble reactive P at a mean influent concentration of 21.0±2.9 mg/l. Overall, the system holds great promise as a novel CWs for simultaneous removal of P and OM, and at the same time, it transforms alum sludge from a waste into a useful material.

Keywords: Alum sludge, constructed wetlands, phosphorus, tidal flow, self organizing maps
1. INTRODUCTION

Constructed wetland systems (CWs) are wastewater treatment systems that encompass a plurality of treatment modules which are akin to processes occurring in natural wetlands. They have the advantage of low cost and lower energy consumption and they have been applied globally to treat various types of wastewaters (Babatunde et al. 2008; Park, 2009). Generally, their performance is good in terms of removal of suspended solids (SS) and organic matter (OM), but inconsistent and often low for nutrient reduction (particularly phosphorus (P)). Consequently, CWs research has been geared towards improving P removal using novel materials with high P adsorption capacity (Johansson Westholm, 2006, Babatunde et al., 2009). Our previous study has demonstrated the feasibility of using dewatered alum sludge cakes as main substrate in a single-stage model CWs to enhance P removal (Zhao et al., 2009). The current study is concerned with the design and performance of a multistage model of the CWs employing dewatered alum sludge as major substrate. The system has two unique features in its design: (i) Use of dewatered alum sludge cakes as main substrate to enhance P removal and for biofilm attachment. Alum sludge is the most widely generated water treatment residual worldwide, and is mostly landfilled since it is regarded as a by-product of little known reuse value (Babatunde and Zhao, 2007). However, the chemical composition of alum sludge, particularly its abundant aluminium gives it a highly reactive surface and a strong affinity for P immobilization (Makris et al., 2005; Makris and O’Connor, 2007; Babatunde et al., 2009), and it can also serve as biofilm carrier (Zhao et al., 2009). Its use as a CWs medium has the potential of improving wastewater treatment and also transforming alum sludge from ‘waste’ into useful material; (ii) Use of multiple CWs stages operated with tidal flow strategy to better simulate the practical use of this novel CWs system. Tidal flow operation is a batch wise, fill-and-draw type operation (Green et al., 1997, 1998; Sun et al., 1999). By operating multiple CWs stages in series using the tidal flow mode, the capacity for oxygen transfer is increased leading to a greater organic matter mineralization.
2. MATERIALS AND METHODS

The CWs consist of 4 interlinked stages set up in the laboratory using Pyrex columns with internal diameter of 9.5 cm. Each stage was filled up to a depth of 50 cm with air-dried dewatered alum sludge cakes which were collected from a local water treatment plant (Zhao et al., 2009). After collection, the sludge was air-dried, ground and sieved to have a d$_{10}$ and d$_{60}$ of 0.5 mm and 1.8 mm, respectively (Babatunde et al., 2009). Each stage had 10 cm of 6-10 mm gravel at the base to serve as support and young *Phragmites australis* was planted on top of each stage. The CWs was operated using the tidal flow strategy which was carried out in cycles with a hydraulic loading rate (HLR) of 1.27 m$^3$/m$^2$.d. Each cycle consist of 1 hour of wastewater contact and 3 hours of stage resting. The influent wastewater had a concentration of 392.7 ± 95.6 mg/l (BOD$_5$), 579.8 ± 142.0 mg/l (COD), 45.2 ± 6.2 mg-P/l (RP (reactive P), which refers to P determination carried out on unfiltered samples), 21.0 ± 2.9 mg-P/l (SRP (soluble reactive P), which refers to P determination carried out on filtered samples), 218 ± 97.8 mg/l (SS) and 7.81 ± 0.25 (pH). Influent and effluent samples from the system were collected periodically and analyzed for BOD$_5$, COD, SS, turbidity, P-PO$_4^{3-}$ (both RP and SRP), pH, NH$_4$-N, NO$_2$-N, NO$_3$-N and TN according to standard methods (APHA, 1998). Self organizing maps (SOM) technique (Aguado et al., 2008) was used to mine data on BOD$_5$, COD, RP and SRP. Dissolved oxygen profile was also monitored in the headspace of stages 1 and 2 of the CWs in order to examine the tidal flow concept. The theoretical amount of oxygen drawn into each stage of the system in each tidal cycle was also determined. The determination is however a simple and basic approach meant as a guide only as removal of OM is quite a complicated and extremely biological process which can also be influenced by OM adsorption, roots aeration, nitrification/denitrification, or anoxic removal of OM.

3. RESULTS AND DISCUSSION
3.1 General treatment performance

Fig. 1 shows the removal trend for organics (BOD₅ and COD) and P (RP and SRP). The system achieved higher removal efficiencies for BOD₅ than for COD. Average removal efficiencies of 90.6 ± 7.5% and 71.8 ± 10.2% for BOD₅ and COD respectively, were achieved. This is quite possible as the ratio of BOD₅ concentration to COD concentration (C_{BOD}/C_{COD}) ranged between 0.59–0.91, which indicates high biodegradability. However, the average C_{BOD}/C_{COD} in the effluent exiting the system was 0.23 indicating low biodegradability. A conceptual model to describe the biodegradation process has been developed (Babatunde et al., 2007). Comparatively, the system achieved a comparable performance with other similar systems such as those reported by Sun et al. (1999); Cerezo et al. (2001) and Zhao et al. (2004a) even though it had a relatively smaller footprint and higher loading. P removal in the system was exceptional. In particular, the system proved very effective for the removal of soluble reactive phosphorus (SRP) and showed superior P removal performance when compared to similar performances in literature such as Sun et al. (1999); Cerezo et al. (2001) and Zhao et al. (2004b).

[INSERT FIG 1 HERE]

The SOM results are shown in Fig. 2. From the mapping, it can be determined that the BOD-in component is mostly negatively correlated with the BOD-out component. This indicates that even with higher influent BOD values, the effluent BOD values are still low and further suggests that in the CWs under study, it will be possible to obtain higher performance and achieve low effluent BOD₅ values even at high influent values. For the COD mapping, the lower part of the COD-in map only had some slight positive correlation with high COD-out values. This is because that the COD in the final effluent from the system was relatively higher that the BOD₅ values. As it has been discussed previously, average BOD₅/COD value of final effluent was 0.23, indicating that most of the organics in the wastewater have been
degraded in the CWs. For RP and SRP, the local average values for both RP-out and SRP-out were very low irrespective of the influent levels. The exceptional ability of the system to achieve very low effluent P concentrations irrespective of the influent P concentration can be attributed to the P adsorption ability of the alum sludge (Yang et al., 2006a, b; Babatunde et al., 2009).

[INSERT FIG. 2 HERE]

The removal fraction in each stage of the system is shown in Table 1. High removal fractions were obtained in the first stage of the system irrespective of the pollutant and this reflects its key role. The individual contribution of the subsequent stages to overall removal ranged from 6-23% (BOD₅) and 7-16% (COD). Nonetheless, the reduction obtained in stage 4 had some contribution to the overall reduction. However, there was no significant P removal beyond the first stage. P removal will be significantly performed in subsequent stages when the first stage becomes saturated. The removal of SS and the reduction of turbidity followed the same trend as the removal of organics with the highest and most significant reduction obtained in the first stage, while although there were further reductions in the subsequent stages, their relative contribution were less significant. It is however worthy to note that effluent ponding on the surface was experienced but this was resolved by bed resting.

There was a slight reduction in the overall removal of total nitrogen (TN), with an average overall removal of 22.8% obtained. However, NH₄-N was reasonably removed with an average overall removal of 73.2% obtained. Furthermore, there was an increase in the level of NO₃-N which suggests nitrification as the nitrifying bacteria converted NH₄–N in the influent to NO₂–N and further to NO₃–N.

[INSERT TABLE 1 HERE]
3.2 Dissolved oxygen profiling

The results (data not shown) of the oxygen profiling show that the percentage decrease in oxygen was higher in stage 1 than in stage 2, indicating that more dissolved oxygen is utilised in the first stage than in the second stage. Furthermore, based on the trend in the percentage oxygen concentration, the following suggestions can be made. Prior to pumping of wastewater, the headspaces in the stages were equilibrated with the atmosphere and therefore saturated with air. During and after the pumping which is also the period of contact of the system with wastewater, there was a little drop in the oxygen concentration in the system but this could have been caused by air-drift, instrument error, or it is possible that actual microbial-degradation is taking place. The change noticed during the pump out stage is probably due to suction effect of the draining wastewater. However, the most significant drop in concentration was when the stages were at rest and the decrease was calculated to be ca.16% and 10% in the first and second stages respectively. A 16% change would imply that 40.32 mg/l of oxygen was utilized by the microorganisms in the first stage and up to 80.64 mg/l of BOD$_5$ can be removed in the first stage. Consequently, 10% - 25% of the oxygen input is available as dissolved oxygen for microbial oxidation and this puts the oxygen transfer efficiency of the system under study at about 5.3-13.3 g-O$_2$/m$^2$. cycle.

CONCLUSIONS

A novel multi-stage constructed wetland system (CWs) using alum sludge as substrate was operated to enhance concurrent removal of phosphorus (P) and organic matter from wastewater. The system achieved high removal efficiencies for BOD$_5$ (90.6%), COD (71.8%), reactive P (80%) and soluble reactive P (89%). The concept holds great promise as an attractive option for achieving high and concurrent removal of P and OM from wastewaters in CWs and in addition, it borders on sustainability by reusing a by-product as its main substrate.
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References


Table 1 Overall and inter-stage treatment performance in the system

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Influent</th>
<th>Stage 1</th>
<th>Stage 2</th>
<th>Stage 3</th>
<th>Stage 4</th>
<th>% removal</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOD₅</td>
<td>392.7</td>
<td>184.1 (53)</td>
<td>93.7 (23)</td>
<td>60.4 (9)</td>
<td>37.1 (6)</td>
<td>90.6</td>
</tr>
<tr>
<td>COD</td>
<td>579.8</td>
<td>353.7 (39)</td>
<td>260.7 (16)</td>
<td>200.1 (11)</td>
<td>158.1 (7)</td>
<td>71.8</td>
</tr>
<tr>
<td>SRP</td>
<td>21</td>
<td>2.3 (89)</td>
<td>1.2 (5)</td>
<td>0.86 (2)</td>
<td>0.47 (2)</td>
<td>97.6</td>
</tr>
<tr>
<td>RP</td>
<td>45.3</td>
<td>8.9 (80)</td>
<td>5.7 (7)</td>
<td>3.7 (4)</td>
<td>3.0 (2)</td>
<td>93.3</td>
</tr>
<tr>
<td>SS</td>
<td>218</td>
<td>72.9 (67)</td>
<td>39.8 (15)</td>
<td>29.2 (5)</td>
<td>19.3 (5)</td>
<td>89.3</td>
</tr>
<tr>
<td>Turbidity</td>
<td>127.7</td>
<td>46.4 (64)</td>
<td>26.5 (16)</td>
<td>18.6 (6)</td>
<td>14.3 (3)</td>
<td>86.7</td>
</tr>
<tr>
<td>TN</td>
<td>142.5</td>
<td>133.8 (64)</td>
<td>125.6 (6)</td>
<td>120 (4)</td>
<td>110 (7)</td>
<td>22.8</td>
</tr>
<tr>
<td>NH₄-N</td>
<td>132.9</td>
<td>87.1 (35)</td>
<td>46.8 (30)</td>
<td>40.2 (5)</td>
<td>35.6 (4)</td>
<td>73.2</td>
</tr>
<tr>
<td>NO₂-N</td>
<td>0.75</td>
<td>0.23</td>
<td>0.13</td>
<td>0.06</td>
<td>0.15</td>
<td>—</td>
</tr>
<tr>
<td>NO₃-N</td>
<td>3.6</td>
<td>9</td>
<td>27.2</td>
<td>9.5</td>
<td>25</td>
<td>—</td>
</tr>
<tr>
<td>pH</td>
<td>7.81</td>
<td>7.0</td>
<td>6.82</td>
<td>6.82</td>
<td>6.81</td>
<td>—</td>
</tr>
</tbody>
</table>

All parameter values are in mg/l, except pH (no unit) and turbidity which is in NTU. Bold values in bracket refer to the amount of removal in percentage obtained in each respective stage, relative to the initial concentration in the wastewater and expressed as a percentage.
Figure captions:

Figure 1. Trend of pollutant removal efficiencies in the novel constructed wetland system

Figure 2. Abstract visualization of the relationship between influent and effluent values of BOD$_5$, COD, RP and SRP in the constructed wetland system using self organizing maps
Fig. 1
Fig. 2