Applying solubilization treatment to reverse clogging in laboratory-scale vertical flow constructed wetlands

Hua Guofen, Zhu Wei, Zhao Lianfang and Zhang Yunhui

ABSTRACT

Substrate clogging is characterized as a frequently occurring operational problem for subsurface-flow constructed wetlands. The application of solubilization treatment to reduce clogging was tested in lab-scaled setups to provide a promising solution. The performance of solubilization treatment on reducing clogging and the related effects on plants and biofilms in the wetland system were investigated in this paper. The results showed that the infiltration rate and available porosity of wetland substrate increased as a function of increased dosage of NaOH, HCl, NaClO, and detergent, respectively. Among the four solvents, it appeared that NaClO had the most obvious effects on reducing clogging and the infiltration rate and effective porosity recovered to 69% of the original condition. The two possible reasons for solubilization were the flocculents’ structure of the clogs was broken up or parts of the organic clogs were dissolved. The function of adding NaOH and NaClO was to dissolve the protein and polysaccharides of the organic clogs; the function of adding HCl was to release the anaerobic gas wrapped in the organic clogs. Furthermore, experiments results also showed that the solubilized solvents did not demonstrate a long-term negative effect on plants and biofilms.

Key words | clogging, constructed wetlands, restore cycle, solubilization treatment

INTRODUCTION

Among several technologies applied to treat wastewater, especially decentralized wastewater, subsurface-flow constructed wetlands is a popular alternative because of its natural-oriented concept and its low cost (Chiarawatchai & Otterpohl 2008).

One major operational problem for subsurface-flow constructed wetlands is clogging, as has been pointed out by several studies (Platzer & Mauch 1997; Pedescoll et al. 2009). Clogging of wetland media will lead to decreased hydraulic conductivity, resulting in problems such as surface flow of wastewater, dead zones, or short circuits (Pedescoll et al. 2009). These problems would negatively affect the general treatment performance as well as its operational lifespan (Langergraber et al. 2003; Chiarawatchai & Otterpohl 2008). Clogging is a complex process, and the reasons for substrate clogging include accumulation of suspended solids, surplus sludge production, chemical precipitation, and deposition in the substrate pores, growth of plant-rhizomes and roots, generation of gas, and compaction of the clogging layer. This issue is widely discussed in previous work (Cooper & Green 1995; Langergraber et al. 2003; Davison et al. 2005; Caselles-Osorio & García 2006; Caselles-Osorio et al. 2007)

Usually, substrate clogging occurs because of the accumulation of solid content in wastewater on the surface or within the substrate layer of vertical-subsurface-flow constructed wetlands (VSFCWs) and at the inlet of horizontal-subsurface-flow constructed wetlands (HSFCWs) (Blazejewski & Murat-Blazejewska 1997; Langergraber et al. 2003; Caselles-Osorio & García 2007). Generally, the clogging problem can be dealt with by two ways: one is to take precautions (such as septic tank and...
reported two case studies of remediation of full-scale subsurface-flow treatment wetlands by a very strong oxidant-concentrated hydrogen peroxide (35%), which is capable of oxidizing even nonbiodegradable components of biofilms.

In this paper, we have investigated in situ application of solubilization treatment to restore hydraulic conductivity of clogged gravel beds in a lab scale. We make attempt to find a solvent that can dissolve or disperse the clogging deposits based on the oxidation, reduction, and acid and alkaline properties and then make them flow out with the wastewater, substantially reducing the clog without destroying or stopping the wetland’s system; this is called solubilization treatment. Because the renovation of wetland nutrients depends on microbial biomass and plants, hydraulic conductivity should be recovered without long-term adverse effects on wetland microorganisms and plants. The restoration cycle of biofilms and plants has been investigated.

MATERIALS AND METHODS

Experimental setups (or strategy)

The experimental system consisted of five identical beds made of Perspex columns 200 mm in height and 110 mm in diameter. The wetland cell was covered by the wetland plant Typha latifolia and filled with pea gravel ($d_{10} = 2.1$ mm; uniformity coefficient $= 2.5$; infiltration rate $= 3.85 \times 10^{-2}$ cm s$^{-1}$, total porosity $= 31.50\%$). The wastewater from the surface entered the wetland cell and discharged from the bottom. Figure 1 shows a schematic of the wetland cell. Because clogging occurred in the substrate’s upper layer (10–20 cm), the height and diameter of the wetland cell correspond to the depth of the clogging layer and the diameter of plants in a cluster density, which means that the entire simulator is equivalent to the interception of the actual clogged wetlands.

It has been reported (Nguyen 2000; Caselles-Osorio et al. 2007) that substrate clogging in constructed wetlands is caused by a mixture of organic and inorganic particles and microorganisms embedded in an organic matrix of bacterial exopolymers form a high moisture, low-density
colloidal sludge similar to activated sludge that comes from municipal wastewater treatment plants. Therefore, in the lab, the activated sludge was used as the simulated clogging matter to shorten the clogging time. The substrate and the activated sludge, which came from municipal wastewater treatment plant of Nanjing city in Jiangsu Province of China, were mixed until the effective porosity was 5.3%, which is when the clogging occurred.

Four different types of solvents (Zhu et al. 2009)—alkali (sodium hydroxide 5.0 g L\(^{-1}\)), acid (hydrochloric acid 5.0 ml L\(^{-1}\)), oxidants (sodium hypochlorite 5.0 ml L\(^{-1}\)), and detergent (Diao Brand, made in China) (5.0 g L\(^{-1}\))—were applied into the wetlands unit model where the clogging had occurred. The substrate had been covered by the solvents for about 8 hours and then the solvents were discharged. The application procedure was repeated daily until the effective pore volume and the infiltration rate measured became stable.

**Physical-chemical measurements**

Organic blocking morphology was observed through an Olympus microscope. Organic blocking particle size distribution was analyzed by an automatic laser particle size analyzer (type LS13320). Chemical analyses of influent and effluent concentrations were performed on conventional methods. Chemical oxygen demand used a rapid airtight catalysis method, including the spectrophotometric method, and the measurement used was the Water Quality Objectives for National Standard of China (Water and Wastewater Monitoring and Analysis Methods 2002). Polysaccharide was determined by the anthrone-sulfuric acid method; for protein, we used Coomassie Brilliant Blue G-250 colorimetry, and for DNA, we used a modified...
diphenylamine method. The specific steps are detailed in accordance with the Biochemistry Experiment (Peng et al. 1989).

Microbiological measurements

To investigate whether there was a long-term negative impact on biofilms, after solubilization treatment each substrate sample was taken on a regular schedule and microbial quantity and enzymatic tests were performed to characterize the activity of the microbial community. Bacteria, actinomycetes, nitrifying bacteria, and nitrogen-fixing bacteria count used coated plate counting; the detection of urease activity of substrate was accomplished by Nessler’s colorimetric method; and sucrase was determined with 3,5-dinitrosalicylic acid (DNS) colorimetric method. All the specific steps are detailed in accordance with the Microbial Analysis Methods Manual (Xu & Zheng 1986).

RESULTS AND DISCUSSION

The performance of solubilization treatment to reduce clogging

The degree of clogging can be characterized by the infiltration rate and the effective porosity. If the clogging can be reduced by solubilization treatment, the infiltration rate and the effective porosity will rise.

From Figures 2 and 3, it can be concluded that the effective porosity rate increased in various degrees by adding four different solvents compared with tap water, from about 5% to 18%. The effect of NaClO treatment was the most obvious, increasing to 22.91%, which was close to the initial effective porosity (27.39%). In addition, stability of effective porosity was achieved for 5 days by adding sodium hypochlorite solution; the remaining solution became stable in 7 days. Similar results were obtained on the infiltration rate measurements. These increased from $3.62 \times 10^{-3}$ cm s$^{-1}$ to about $2.4 \times 10^{-2}$ cm s$^{-1}$. The most obvious effect of sodium chlorate increased to $2.69 \times 10^{-2}$ cm s$^{-1}$ and recovered to the initial value of 69%, which is similar to conclusions drawn by Shaw (Magesan et al. 2000). Because of the large amount of foam in water

Plant measurements

For the same purpose as the microbiological measurements, the plant root activity, soluble sugar, and catalase ($H_2O_2$) were measured every 7 days. For assays of the soluble sugar content, the anthrone-colorimetric technique was adopted. Catalase was measured by ultraviolet absorption method; the root activity was expressed with triphenyl tetrazolium chloride (TTC) reduction intensity, which was found in the Principle and Technology on Physiological and Biochemical of Plant. (Li et al. 1999).
caused by adding detergent, the detergent was not further investigated as an available solvent.

The reasons for solubilization treatment to reduce clogging

It was hypothesized that the clogs attached on the substrate could be removed by dissolving them or breaking them into small particles and discharging them with effluent. The microscope pictures enlarged 400 times in Figure 4 show that the structure of the clogging matter is different after solvent solubilization treatments. Figure 5 describes the size distribution of treated clogs after solubilization treatment.

As can be seen from Figure 4, the blocking compound was dispersed less effectively by hydrochloric acid treatment compared with NaOH and NaClO treatments. Parts of the flocculent structure were destroyed and even the microbial cell structure was broken by the NaClO treatment.

As can be seen from Figure 5, the particle size after solubilization treatment became smaller especially after treatment by the NaOH and NaClO solvents compared with tap water. It was shown that \( d_{50} \) could change from 140.2 \( \mu \)m into 36.2 \( \mu \)m and 38.4 \( \mu \)m, respectively. Therefore, it could be speculated that not water, but acidic or alkaline substances or oxidation of material or another substance, played an important role in the reduced clogging.

So COD, DNA, polysaccharide, and protein of clogs were tested to explore what was dissolved out after different solubilization treatments. Analytical results after solubilization treatment are presented in Table 1.

There were differences in dissolving the organic matter among the three solubilization treatments. The most significant gaps were 5.2 times and 32.7 times the dissolution of polysaccharide and protein after NaClO and NaOH treatment, respectively. The solubilization treatments could dissolve the exopolysaccharide biofilm matrix and restore permeability to the plugged cores. The DNA was dissolved by NaClO treatment, which meant that it would damage the cells to a certain extent. The dissolution of organic matter by HCl treatment was minor and close to the initial values.

However, what was dissolved by HCl treatment? Why was the clogging substantially reduced? When HCl was added to the organic blocking, a large amount of bubbles was observed and some gases, which smelled like rotten eggs. To clarify the composition of the produced gas, a wet starch-potassium iodide paper was applied, which identified the produced gas as \( H_2S \). Then the volume 0.35 ml g\(^{-1}\) was quantitatively measured by the spectrophotometric methylene blue method. The gases produced by sulfur-reducing bacteria and methanogenic bacteria in the wetlands formed the saturated zone, which may be one of the reasons for congestion (Leverenz et al. 2009). The possible reason for solving the clogging problem by HCl was that: the gas produced by the reaction with HCl and the anaerobic gases blocked in the pores were released.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Different item contents of the solubilization</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Polysaccharide mg L(^{-1})</td>
</tr>
<tr>
<td>Initial values</td>
<td>10.53</td>
</tr>
<tr>
<td>HCl</td>
<td>16.02</td>
</tr>
<tr>
<td>NaOH</td>
<td>46.02</td>
</tr>
<tr>
<td>NaClO</td>
<td>55.03</td>
</tr>
</tbody>
</table>

Figure 5 | Particle size distribution of treated clogs after solubilization treatment.
The impact on wetlands system after solubilization treatment

The impact on wetlands biofilms

Micro-organisms are the main force of sewage purification in constructed wetlands (U.S. EPA 2000; Tietz et al. 2008). To a large extent, the number of micro-organisms represents the wastewater purifying capacity. The changes in the number of bacteria, actinomycetes, nitrifying bacteria, and nitrogen-fixing bacteria on the first and seventh days after solubilization treatment could repair the damage to the micro-organisms and restore the cycle. Experimental results are shown in Table 2.

Table 2 shows that different types of solvents from the dissolution of micro-organisms produced different degrees of damage to the micro-organisms. The number of bacteria, actinomycetes, nitrifying bacteria, and nitrogen-fixing bacteria on the first and seventh days after solubilization treatment could repair the damage to the micro-organisms and restore the cycle. Experimental results are shown in Table 2.

Table 2 shows that different types of solvents from the dissolution of micro-organisms produced different degrees of damage to the micro-organisms. The number of bacteria, actinomycetes, nitrifying bacteria, and nitrogen-fixing bacteria on the first and seventh days after solubilization treatment could repair the damage to the micro-organisms and restore the cycle. Experimental results are shown in Table 2.

Table 2 | Different amount of microorganisms after solubilization treatment

<table>
<thead>
<tr>
<th></th>
<th>NaOH</th>
<th>HCl</th>
<th>NaClO</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Initial values</td>
<td>1st day</td>
<td>7th day</td>
</tr>
<tr>
<td>Bacterial</td>
<td>$8.32 \times 10^6$</td>
<td>$8.01 \times 10^5$</td>
<td>$6.21 \times 10^5$</td>
</tr>
<tr>
<td>Actinomycetes</td>
<td>$5.08 \times 10^4$</td>
<td>$6.03 \times 10^3$</td>
<td>$6.09 \times 10^3$</td>
</tr>
<tr>
<td>Nitrifying bacteria</td>
<td>$2.01 \times 10^3$</td>
<td>$8$</td>
<td>$6.22 \times 10^2$</td>
</tr>
<tr>
<td>Nitrogen-fixing bacteria</td>
<td>$6.81 \times 10^3$</td>
<td>$11$</td>
<td>$1.02 \times 10^3$</td>
</tr>
</tbody>
</table>

The impact on wetlands plants

Plant roots are the active organs of absorption and synthetic processes and directly respond to the vitality of the plant. The solvent directly contacted with plant roots by using biochemical processes, such as the synthesis and decomposition of humus and organic compounds, turning debris of higher plants and micro-organisms into the form available. There is a significant correlation between urease activity and the removal of Kjeldahl nitrogen (KN) (Liang et al. 2003); sucrose is an important enzyme in the carbon cycle that significantly related to the degradation rate of polysaccharides (Guan 1986).

The changes of sucrose and urease enzyme activity on the first and seventh days after solubilization treatment represent the damage to the activities of micro-organisms and the restored cycle. Experimental results are shown in Table 3.

Table 3 shows that different types of solvents from the dissolution of micro-organisms produced different degrees of damage to the micro-organisms. The number of bacteria, actinomycetes, nitrifying bacteria, and nitrogen-fixing bacteria on the first and seventh days after solubilization treatment could repair the damage to the micro-organisms and restore the cycle. Experimental results are shown in Table 3.

Table 3 | Different enzymatic activities of the substrate after solubilization treatment

<table>
<thead>
<tr>
<th>Enzyme activity</th>
<th>Initial values</th>
<th>NaOH</th>
<th>HCl</th>
<th>NaClO</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1st day</td>
<td>7th day</td>
<td>1st day</td>
<td>7th day</td>
</tr>
<tr>
<td>Sucrase ($\mu$g g$^{-1}$)</td>
<td>12.05</td>
<td>1.08</td>
<td>11.21</td>
<td>0.15</td>
</tr>
<tr>
<td>Urease ($\mu$g g$^{-1}$)</td>
<td>3.86</td>
<td>1.12</td>
<td>3.05</td>
<td>0.98</td>
</tr>
</tbody>
</table>
solubilization treatment, which is bound to affect the growth of the plants. Therefore, measuring the activity of plants roots can indicate the impact of the solvent on the plants. The phenomenon of plant growth after solubilization treatment is shown in Figure 6 and the results are shown in Table 4.

As can be seen from the Figure 6, root activity dramatically reduced, and some even could not be measured on the first day after solubilization treatment. However, even in this case, root activity was restored on the 28th day after HCl and NaOH treatment. That the root of the plants had been filled with the porosity of gravel after HCl treatment indicated that the plants grew well, whereas plant roots had been partially decomposed after NaClO treatment, as observed in Figure 6. As a consequence of strong oxidation and alkaline nature of NaClO, the damage to the plant root system is relatively extensive. Future experiments should focus on optimizing the residence time and the concentration of NaClO to reduce the plant damage.

Because the root vigor was determined by destructive testing, periodic results were simply known and the process of the impact on the plant was not clear. After solubilization treatment, every 7 days the soluble sugar and catalase of the plants were measured to reflect the recovery cycle of plants. The results are shown in Figure 7.

Table 4 | Different activities of plant roots after solubilization treatment unit (μg g⁻¹ h⁻¹)

<table>
<thead>
<tr>
<th></th>
<th>HCl 1st day</th>
<th>HCl 28th day</th>
<th>NaOH 1st day</th>
<th>NaOH 28th day</th>
<th>NaClO 1st day</th>
<th>NaClO 28th day</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>29.02</td>
<td>59.51</td>
<td>–</td>
<td>42.64</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

–, Not detected.
In the environment stress process after solubilization treatment, the fluctuations of soluble sugar and catalase took place as a result of a stress response to the solvent. However, after about 15 days, these fluctuations became stable when the plants returned to normal. From the analysis presented here, it appears that the solvent did not have a long-term negative effect on the plants and biofilms after solubilization treatment.

**CONCLUSIONS**

Solubilization treatment is considered a promising method to remediate clogging in subsurface-flow treatment wetlands. This paper presented the remediation of clogging by adding three solvents (HCl, NaOH, and NaClO) in low concentrations into a vertical-subsurface-flow constructed wetland (VSSF) unit model. The effective porosity and infiltration rate were improved and the clogging was reduced.

The reasons for the solubilization treatment to reduce clogging were categorized into two aspects and were not exactly the same for the three different solvents. The blocking compound was dispersed and parts of the flocculent structure were destroyed by the solubilization treatment, which broke up the clogging materials that were discharged with the waste water. Proteins and polysaccharides were mainly dissolved by the NaOH and NaClO treatment and the generated anaerobic gas packed in the pore space was possibly released by HCl treatment.

Clogging was substantially reduced and the solvent did not demonstrate a long-term negative effect on plants and biofilms after solubilization treatment. Future research in this area should focus on optimizing the amount of solvent dosage, which can effectively treat clogged vertical-subsurface-flow wetlands with restricted damage to the plants and biofilms.

Furthermore, it should be pointed out that there is some potential gap between lab-scale experiments solutions found by this paper for the limited frame and sites in operation. The presented solution has to be validated at a larger scale.

**ACKNOWLEDGEMENTS**

The authors are grateful to the financial supports for the research Natural Science Foundation of Jiangsu Province (No.BK2006710). Also, sincere appreciation has to be given to all of those who provided the valuable information. Without them, this paper would not have fulfilled its objective.

**REFERENCES**


