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Removal of Ammoniacal-Nitrogen Removal From an Artificial Landfill Leachate in Downflow Reed Beds

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Abstract

The fate of ammoniacal-nitrogen ($\text{NH}_4\text{-N}$) was studied in a lab-scale downflow reed bed system treating an artificial landfill leachate. Individual reed beds were submerged by the leachate, then drained and rinsed by tap water. It was discovered that $\text{NH}_4\text{-N}$ was removed by a two-staged process, adsorption onto the reed bed media followed by nitrification into nitrite-nitrogen ($\text{NO}_2\text{-N}$) and nitrate-nitrogen ($\text{NO}_3\text{-N}$). A drop of $\text{NH}_4\text{-N}$ level of the leachate was observed when the reed beds were submerged. By rinsing of the beds, part of the $\text{NH}_4\text{-N}$ adsorbed inside the bed matrices was re-released into the rinse water. The presence of $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ in the rinse water demonstrated that nitrification process took place while the $\text{NH}_4\text{-N}$ was being retained inside the bed matrices. For artificial leachates with $\text{NH}_4\text{-N}$ levels of 150 ± 5 mg/l, an average removal rate of 43.8% in a three-hour treatment was achieved; mass balance analysis indicated that processes of adsorption, and transformation into $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ accounted for 63.7%, 4.3% and 24.4% of the $\text{NH}_4\text{-N}$ removal, respectively. This study also demonstrated that in general greater recirculation rate of effluent around the downflow reed beds gives higher $\text{NH}_4\text{-N}$ removal.

Keywords: Adsorption; ammoniacal-nitrogen; leachate; nitrification; reed bed; wastewater treatment.

Introduction

Landfill leachates are difficult to characterise because their compositions and concentrations depend on a variety of factors, such as waste composition, age of landfill site, geology, temperature, moisture content and other seasonal and hydrological factors (Im *et al.*, 2001). It is generally recognised that mature leachates contain relatively low concentrations of degradable organic material but high levels of $\text{NH}_4\text{-N}$, up to 5000 mg/L (Knox, 1985; Li *et al.*, 1999). A high level of $\text{NH}_4\text{-N}$ can inhibit the microbial activity in biological treatment processes and negatively affect the capacity of the treatment system (Robinson *et al.*, 1992; Li and Zhao, 2001). Conventionally, landfill leachates are transported to wastewater treatment facilities where they are treated along with domestic sewages. However, various hazardous substances in the leachates can jeopardise biological processes of the treatment. In addition, transportation of leachates is often costly and raises concerns for public safety. Hence, an effective and low-cost on-site treatment technique appears to be the ideal solution for leachate problems.

Reed bed technology works on the principle that reeds have the ability to transfer a certain amount of oxygen to their roots, stimulating microbial activities and breaking down pollutants in the root zone (Robinson *et al.*, 1992). As a 'green' technology with low energy consumption, reed bed systems have become one of the most popular wastewater treatment alternatives in many areas of the world, especially in countries where commitments were made in Kyoto 1997 to cut emissions of greenhouse gases by at least 5 per cent by year 2012. For the treatment of landfill leachates reed bed systems have distinctive advantages and are pre-competitive because of their efficiency and low operational cost. Landfill sites can usually provide the necessary space for constructing reed beds, while reeds have an aesthetical appearance which compensates for the poor image of the sites. In recent years there has been an increasing interest in the

application of reed beds in landfill leachate treatment (Sanford *et al.*, 1995; Bulc *et al.*, 1997; Martin *et al.*, 1999).

Compared with the removal of organic matter, $\text{NH}_4\text{-N}$ is more difficult to remove in reed beds as nitrification bacteria are autotrophic microorganisms that have a slow respiration rate and require a considerable amount of oxygen to function. With respect to the nitrification of $\text{NH}_4\text{-N}$, Sun *et al.* (1999) reported that the reduction of $\text{NH}_4\text{-N}$ in a downflow reed bed is not balanced by increases in $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ contents in the wastewater. A similar result was reported by Tyrrel *et al.* (2002) that the mass of $\text{NO}_3\text{-N}$ present in treated leachate did not account for the mass of $\text{NH}_4\text{-N}$ and $\text{NO}_2\text{-N}$ being removed. Both studies proclaimed the ‘disappearance’ of $\text{NH}_4\text{-N}$ might be a result of a combination of different processes, which involved plant uptake, adsorption, volatilisation, etc. However, the exact nature and mechanisms of these processes are still unclear. In this study, laboratory experiments were designed to treat an artificial landfill leachate using a gravel-based vertical downflow reed bed system that was operated in batch and continuous modes with different ratios of effluent recirculation to raw material flow rates. A special focus was placed on the fate of $\text{NH}_4\text{-N}$ removed in the system.

Materials and Methods

An artificial leachate was synthesized to simulate weak methanogenic landfill leachates using di-ammonium hydrogen orthophosphate ($(\text{NH}_4)_2\text{HPO}_4$), tap water and a small amount of caster sugar. Table 1 shows the properties of the artificial leachate. ‘Aged leachate’ in Table 1 refers to the leachate that was stored in a storage tank for two weeks before experiments.

Insert Table 1

The lab-scale reed bed system consists of two identical beds that were made of Perspex columns of 88cm in height and 10cm internal diameter. Each bed was filled with 20-30 mm round gravel to a depth of 20 cm and a top layer of 2-5 mm grit with a depth of 60 cm. A single common reed, *Phragmites australis*, was planted in the top layer of each bed. Figure 1 shows a schematic diagram of the reed bed system.

Insert Figure 1

To prevent excessive build-up of biomass the reed beds were operated alternately, one operating while the other is resting. Experiments were carried out in two periods. During Period 1 (batch experiment), the operating reed bed was submerged by both ‘aged’ and fresh artificial leachate for three hours while small amounts of samples were taken from the bottom of the bed every half hour. The use of ‘aged’ leachate was to test the effect of an initial build-up of nitrification bacteria on $\text{NH}_4\text{-N}$ removal in the beds, whereas the use of freshly prepared artificial leachate was to prevent fluctuations in $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ levels and provide more precise control for the experiments. For the fresh leachate a rinse experiment was also carried out; after three hours of submerging the operating bed was drained, left for one hour for natural aeration then rinsed three times with tap water. The rinse waters were then collected, mixed and analysed as one sample. This operation of submerging, draining and rinsing was repeated for five runs when the intervals for natural aeration was increased to two, three, four, five and twenty-five hours before the bed was rinsed.

During Period 2 (continuous experiment), artificial leachate was conveyed to the operating reed bed from a 125 L feed tank using a feed pump (Watson Marlow peristaltic pump, 101U/R Series), as shown in Figure 1. Effluent out of the bed was collected by a flask, where a certain

amount of the effluent was recirculated back onto the reed bed surface and the rest overflowed into an effluent storage tank. Samples were collected from the inlet and outlet of the bed. Daily flow rate of the raw material was set at 5 l/d, and the ratio of recirculation to raw material flow rates was varied according to experiment needs. At each recirculation ratio the system was allowed to stabilize for one day before samples were collected. All wastewater samples collected during experiments were analyzed for $\text{NH}_4\text{-N}$, $\text{NO}_2\text{-N}$, $\text{NO}_3\text{-N}$ and pH.

Results

Period 1

Figure 2 illustrates the average effluent $\text{NH}_4\text{-N}$ levels when the reed beds were submerged and drained with ‘aged’ and fresh artificial leachates. As shown in Figure 2, the bulk removal of $\text{NH}_4\text{-N}$ from the leachates took place fairly quickly, within the first half hour. After half hour, there was no significant change in effluent $\text{NH}_4\text{-N}$ concentration, suggesting that virtually no physical, chemical or biological processes occurred in the bed that separate $\text{NH}_4\text{-N}$ from the artificial leachates.

Insert Figure 2

Average $\text{NH}_4\text{-N}$ concentrations of the ‘aged’ and fresh artificial leachates before and after the batch treatment in the reed beds are presented in Table 2. It was found that for the ‘aged’ leachate, average removal of $\text{NH}_4\text{-N}$ by 33.7% from 148.7 mg/l to 98.6 mg/l occurred when the reed bed was submerged, whereas for the freshly made leachate average $\text{NH}_4\text{-N}$ removal was 43.8%, from 155.9 mg/l to 87.6 mg/l. The reduction of $\text{NH}_4\text{-N}$ for fresh leachate was slightly higher than for the ‘aged’ leachate, suggesting that the storage of leachate and initial build-up of

nitrifying bacteria in the storage tank did not help $\text{NH}_4\text{-N}$ reduction, and the majority of nitrifying bacteria may be present in biofilms attached on surfaces inside the reed bed matrix.

Insert Table 2

In order to discover the fate of $\text{NH}_4\text{-N}$ that was separated and removed from the fresh leachate, a mass balance was conducted using concentrations of $\text{NH}_4\text{-N}$, $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ of the samples. Figure 3 gives results of the nitrogen mass balance in the submerging, draining and rinsing experiments. It can be observed from Figure 3 that decrease in $\text{NH}_4\text{-N}$ corresponds to increases in $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ as a certain amount of $\text{NH}_4\text{-N}$ was nitrified. The mass balance reveals the fate of $\text{NH}_4\text{-N}$ that was separated from the artificial leachate in the reed beds during the submerging process. As illustrated in Figure 4, the removal of $\text{NH}_4\text{-N}$ from fresh artificial leachate is mainly attributed to adsorption by the media (63.7%), and transformation into $\text{NO}_2\text{-N}$ (4.3%) and $\text{NO}_3\text{-N}$ (24.4%).

Insert Figure 3

Insert Figure 4

After the reed beds were submerged and drained with fresh artificial leachate, the matrices were left for natural aeration for 1, 2, 3, 4, 5, and 24 hours before being rinsed with tap water. A considerable level of $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ was found in the rinse water, indicating that a certain amount of $\text{NH}_4\text{-N}$ being adsorbed in the bed matrices was nitrified during the aeration period. Figure 5 presents relationship between the amount of $\text{NO}_3\text{-N}$ and $\text{NO}_2\text{-N}$ generated inside the

reed bed matrices and the aeration time. As shown in Figure 5, the mass of $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ increased as the beds were left for longer periods.

Insert Figure 5

Period 2

The results of continuous experiments with effluent recirculation are presented in Figure 6 and Figure 7. These experiments were designed to investigate the effect of effluent recirculation on $\text{NH}_4\text{-N}$ removal. With recirculation ratios in the range of 0:1 to 10:1 (recirculation flow rate : raw material flow rate) it appears that increases in recirculation ratios lead to higher $\text{NH}_4\text{-N}$ percentage removals although there is a considerable scatter of the data, as shown in Figure 6. Figure 7 indicates that at higher recirculation ratios there are higher net increase rates in $\text{NO}_3\text{-N}$ level (defined as increase in $\text{NO}_3\text{-N}$ concentration divided by its original value), suggesting that the greater $\text{NH}_4\text{-N}$ removal percentages at higher recirculation ratios are a result of enhanced nitrification process. Recirculating effluent around the downflow reed beds gives greater residence time of the artificial leachates in the bed matrices, as well as improved aeration and wastewater-microorganisms interactions, thereby creating a more favourable condition for the activities of nitrifying bacteria.

Insert Figure 6

Insert Figure 7

Discussion

Results from experiments in Period 1 demonstrate that gravel-based downflow reed beds can provide significant removal of $\text{NH}_4\text{-N}$. As the reed beds were submerged, the levels of $\text{NH}_4\text{-N}$ in the ‘aged’ and fresh artificial leachates were reduced by 33.7% and 43.8%, respectively. Surprisingly, it was found that the removal of $\text{NH}_4\text{-N}$ from the leachates happened fairly quickly. The question arises how the $\text{NH}_4\text{-N}$ was removed. Was $\text{NH}_4\text{-N}$ instantly transformed into $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ as the reed beds were being submerged?

Nitrification of $\text{NH}_4\text{-N}$ by microorganisms is normally a slow process as the autotrophic nitrifying bacteria have a low respiration rate. The rapid removal of $\text{NH}_4\text{-N}$ from artificial leachates during the submerging process and the release of $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ into the rinsing water suggest that $\text{NH}_4\text{-N}$ was not transformed biologically into $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ as the beds were being submerged; instead, $\text{NH}_4\text{-N}$ might have been adsorbed and retained inside the reed bed matrices. In contrast to the slow nitrification reaction, adsorption is a rapid and reversible process; this might be the reason that some of the $\text{NH}_4\text{-N}$ was rapidly separated from the leachates and later released as the beds were rinsed by tap water that contained no $\text{NH}_4\text{-N}$. The presence of a considerable level of $\text{NO}_3\text{-N}$ in the rinse water also demonstrated that as $\text{NH}_4\text{-N}$ was being held inside the matrices, a certain amount of the $\text{NH}_4\text{-N}$ was converted into $\text{NO}_2\text{-N}$ then $\text{NO}_3\text{-N}$ by nitrifying bacteria growing in the biofilms attached to the roots of reeds and surfaces of bed media. Therefore, $\text{NH}_4\text{-N}$ was removed in gravel-based downflow reed beds by a two-staged process, adsorption onto the reed bed media followed by nitrification.

With the help of mass balance analysis it was found that for the $\text{NH}_4\text{-N}$ adsorbed and separated from the fresh artificial leachate up to 63.7% was released into the rinse water, 24.4% was transformed into $\text{NO}_3\text{-N}$ and 4.3% converted into $\text{NO}_2\text{-N}$. A small amount, 7.6%, of the $\text{NH}_4\text{-N}$

was unaccounted for and is marked as removed by ‘other’ processes in Figure 4. Apart from nitrification many processes consume $\text{NH}_4\text{-N}$ (Kruzic and Schroedern, 1990; Sun *et al.*, 2002), and these processes, including plant assimilation, ion exchange and combined nitrification-denitrification, etc, may have removed a small amount of $\text{NH}_4\text{-N}$ during the experiments. In addition, it is likely that not all $\text{NH}_4\text{-N}$ being retained inside the bed matrices can be desorbed and released into the rinse water; this may also create a certain amount of $\text{NH}_4\text{-N}$ that seems to have ‘disappeared’. Nevertheless, the bulk of $\text{NH}_4\text{-N}$ separated from the artificial leachate was found in the rinse water, either in the form of soluble $\text{NH}_4\text{-N}$ or transformed into $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$; this further suggests that adsorption followed by nitrification was the main route for $\text{NH}_4\text{-N}$ removal in downflow reed beds.

Figure 5 again indicated that nitrification took place while $\text{NH}_4\text{-N}$ was being retained in the reed beds. After the leachate was released the bed matrices were unsaturated and air diffused from atmosphere into the matrices, providing an adequate oxygen source for the function of nitrifying bacteria attached on surfaces inside the beds. The rate of nitrification may now be controlled by the respiration rate of the microbes. It is inappropriate to speculate the order of nitrification reaction based on results from this study because such kinetics should not be confirmed until substantial data are collected from various systems under different operating conditions.

Figure 6 and Figure 7 suggest that the implementation of effluent recirculation operation in downflow reed beds benefits $\text{NH}_4\text{-N}$ reduction by enhancing nitrification. In current study the increase in $\text{NH}_4\text{-N}$ percentage removal as the recirculation ratios rises from 1:1 to 10:1 may be a result of increased residence time for the leachates, which enables more $\text{NH}_4\text{-N}$ to be nitrified and less nitrifying bacteria to be washed out of the reed beds. Theoretically, recirculating effluent help bacterial growth by increasing retention time and stimulating the reaction of

nitrification-denitrification. Several studies supported this theory. Sun *et al.*, (1998) reported that increased oxygen consumption by microorganisms was achieved when effluent recirculation was employed in a downflow reed bed system treating a mixture of agricultural and domestic effluents. Bahlo (2000) documented a 41% increase in treatment efficiency with a 1:1 recirculation ratio and indicated that greater results may have been achieved had a more favourable $\text{NH}_4\text{-N}:\text{BOD}_5$ ratio been used. However, the benefit of effluent recirculation may not be obvious at the beginning of this operation because multiplication of the population of nitrifying bacteria takes time. Also, $\text{NH}_4\text{-N}$ being adsorbed and retained inside reed bed matrices may create situations that $\text{NH}_4\text{-N}$ is released into the recirculating wastewater; this is believed to be the reason that the data in Figures 6 and 7 are fairly scattered although there is a general trend of increase of percentage $\text{NH}_4\text{-N}$ removal with the recirculation ratio.

It should be pointed out that during the treatment of strong wastewaters in reed bed systems the removal of $\text{NH}_4\text{-N}$ is extremely complicated. It involves a series of physical, chemical and biological processes such as adsorption in the bed matrix, filtration, pollutant sedimentation, ion exchange, plant sorption and microbiological reactions (Brix, 1994). Results from this study do not show a complete picture for the complicated transformations of $\text{NH}_4\text{-N}$. Nevertheless, this study provides useful information to identify the fate of soluble $\text{NH}_4\text{-N}$ removed in downflow reed bed systems, particularly regarding the initial adsorption and retaining of $\text{NH}_4\text{-N}$ inside the bed matrices. Further research work may need to be carried out to establish the equilibrium of $\text{NH}_4\text{-N}$ adsorption and investigate the kinetics of nitrification as $\text{NH}_4\text{-N}$ is adsorbed and retained inside bed matrices.

Treatments of real landfill leachates using reed beds will inevitably require substantial removal of $\text{NH}_4\text{-N}$. Results from this study suggest that to achieve efficient $\text{NH}_4\text{-N}$ removal it is sensible

to maximise contact between the beds media and leachates in order to facilitate $\text{NH}_4\text{-N}$ adsorption, and to provide adequate oxygen to the bed matrix so that the $\text{NH}_4\text{-N}$ adsorbed inside the bed matrix can be nitrified by microorganisms.

Conclusions

Experiment results demonstrate that adsorption plays a key role in the removal of $\text{NH}_4\text{-N}$ from wastewater in gravel-based downflow reed beds. As soon as contact was made between an artificial landfill leachate and media of a reed bed, adsorption of $\text{NH}_4\text{-N}$ inside the bed matrices occurred. A certain amount of $\text{NH}_4\text{-N}$ being adsorbed and retained in the matrices was gradually transformed into $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$. Regarding $\text{NH}_4\text{-N}$ separated from the artificial leachate it was found by a mass balance that on average 24.4% of the $\text{NH}_4\text{-N}$ was transformed into $\text{NO}_3\text{-N}$, 4.3% into $\text{NO}_2\text{-N}$, 7.6% was removed by other processes, whereas the rest of the $\text{NH}_4\text{-N}$ was desorbed and released into the rinse water as the reed beds were washed by tap water. Experiments of continuous treatment of the artificial leachate at various effluent recirculation ratios demonstrated a general increase in percentage $\text{NH}_4\text{-N}$ removal and a rise in $\text{NO}_3\text{-N}$ levels as the recirculation was intensified; this suggested that the operation of effluent recirculation assisted nitrification process.

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Table 1. Properties of artificial leachates used in this study

Experiment	Leachate	NH ₄ -N	NO ₂ -N	NO ₃ -N	PO ₄ -P	COD	pH
Batch	‘Aged’	148.7	9.6	24.4	119.3	27.4	7.2
Batch	Fresh	155.9	1.1	0.5	121.9	22.2	8.1
Continuous	Fresh	122.4	1.3	1.0	125.8	23.6	7.7

Table 2 Average NH₄-N concentrations of the leachate during the batch treatment

	“Aged” leachate	Fresh leachate
Influent, mg/l	148.7	155.9
Effluent, mg/l	98.6	87.6
Percentage removal, %	33.7	43.8

Figure 1 Schematic diagram of the reed bed system

Figure 2 NH₄-N levels as the reed beds were submerged with 'aged' and fresh leachates

Error bars represent: Average $\pm$ SD

Figure 3 Mass balances of NH₄-N, NO₂-N and NO₃-N in batch experiments with fresh leachate

Figure 4 Fate of $\text{NH}_4\text{-N}$ separated from fresh artificial leachate

Figure 5 Amount of $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$ generated vs interval time

Figure 6 Effect of recirculation ratio on $\text{NH}_4\text{-N}$ removal percentage

Figure 7 Effect of recirculation ratio on net NO₃-N increase