Nitrogen removal from concentrated latex wastewater by land treatment

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Abstract

Thongnuekhang, V. and Puetpaiboon, U.
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Songklanakarin J. Sci. Technol., 2004, 26(4) : 521-528

Most of the concentrated latex factories in the South of Thailand discharge treated wastewater that contains high level of nitrogen to a nearby river or canals leading to a water pollution problem. A study of land treatment system was conducted to treat and utilize nitrogen in treated wastewater from the concentrated latex factory. The experimental pilot-scale land treatment system was constructed at the Faculty of Engineering, Prince of Songkla University, Hat Yai Campus. It consisted of water convolvulus (*Ipomea aquatica, I. Reptans*), tropical carpet grass (*Axonopus compresus (Swartz) Beav.*) and control unit (no plantation). The treated wastewater from the stabilization pond system of the selected concentrated latex factory in Songkhla was used to irrigate each experimental unit. Influent and effluent from the experimental units were analyzed for TKN, NH_3-N, Org-N, NO_3^--N, NO_2^--N, BOD_5, sulfate, pH and EC. The land treatment system resulted a high removal efficiency for nitrogen. Tropical carpet grass provided higher removal efficiency than other units for all parameters. The removal efficiency of water convolvulus and control unit were not significantly different. The average removal efficiency of TKN, NH_3-N, Org-N, BOD_5 and sulfate for tropical carpet grass unit were 92, 97, 61, 88 and 52%, for water convolvulus unit were 75, 80, 43, 41 and

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30%, and for control unit were 74, 80, 41, 31 and 28%, respectively. Mass balance of nitrogen transformation was conducted. It revealed that plant uptake was the major mechanism for nitrogen removal in land treatment.

Key words : land treatment, nitrogen mass balance, nitrogen removal, wastewater reuse, wastewater treatment

บทคัดย่อ

วิกาญดา ทองเนื้อแข็ง[1] และ อุดมผล พืชน์ไพบูลย์[2]

การบำบัดไนโตรเจนในน้ำเสียจากโรงงานผลิตน้ำยางข้นด้วยวิธีการบำบัดโดยดิน

ว. สงขลานครินทร์ วทท. 2547 26(4) : 521-528

อุตสาหกรรมน้ำยางข้นเป็นอุตสาหกรรมที่กำลังขยายตัวอย่างมากในภาคใต้ของประเทศไทย   แต่โรงงานอุตสาหกรรมเหล่านี้มักจะปล่อยน้ำทิ้งซึ่งมีไนโตรเจนสูงลงสู่คูคลองและแหล่งน้ำธรรมชาติ ก่อให้เกิดปัญหามลภาวะทางน้ำตามมา จึงได้ทำการศึกษาโดยนำระบบการบำบัดโดยดินร่วมกับพืช มาใช้ในการกำจัดไนโตรเจนพร้อมกับนำไนโตรเจนในน้ำทิ้งจากโรงงานอุตสาหกรรมน้ำยางข้นกลับมาใช้ให้เกิดประโยชน์ โดยได้ทำการสร้างแปลงทดลองขึ้น ณ คณะวิศวกรรมศาสตร์ มหาวิทยาลัยสงขลานครินทร์ วิทยาเขตหาดใหญ่ ซึ่งประกอบด้วย แปลงผักบุ้งจีน หญ้ามาเลเซีย และแปลงควบคุม (ไม่มีการปลูกพืช) และรดน้ำแปลงทดลองแต่ละแปลงด้วยน้ำทิ้งจากโรงงานอุตสาหกรรมน้ำยางข้นในจังหวัดสงขลา ซึ่งเป็นน้ำทิ้งที่ผ่านการบำบัดแล้วด้วยระบบบ่อปรับเสถียร ในการทดลองได้ทำการวิเคราะห์น้ำเข้าและน้ำออกของระบบเพื่อหาค่า TKN, NH_3-N, Org-N, NO_3-N, NO_2-N, BOD_5, Sulfate, pH และ EC จากผลการทดลอง พบว่าระบบนี้มีประสิทธิภาพสูงในการกำจัดไนโตรเจนในน้ำเสีย และพบว่าแปลงหญ้ามาเลเซียมีประสิทธิภาพในการบำบัดน้ำเสียสูงกว่าแปลงทดลองอื่น ๆ   ส่วนแปลงผักบุ้งจีนและแปลงควบคุมนั้นให้ผลที่ไม่แตกต่างกันนัก ประสิทธิภาพเฉลี่ยในการกำจัด TKN, NH_3-N, Org-N, BOD_5 และ Sulfate ของแปลงหญ้ามาเลเซียคือ 92, 97, 61, 88 และ 52% ตามลำดับ สำหรับแปลงผักบุ้งจีนคือ 75, 80, 43, 41 และ 30% ตามลำดับ และสำหรับแปลงควบคุมคือ 74, 80, 41, 31 และ 28% ตามลำดับ และยังได้ทำการศึกษาดุลของไนโตรเจนในระบบ ซึ่งผลการทดลองแสดงให้เห็นว่าการนำไนโตรเจนไปใช้ของพืชเป็นกลไกหลักในการกำจัดไนโตรเจนในน้ำเสียด้วยระบบบำบัดโดยดินร่วมกับพืช

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Land treatment for wastewater has been investigated and practised at several locations in the world for a long time. The advantage of land treatment is the combination of wastewater treatment and recycling in the same process. Wastewater will be applied to the land surface to achieve a specified degree of treatment through the natural physical, chemical, and biological process within the soil-plant-water matrix. This system provides high efficiency for nutrients removal. Boyden and Rababah (1996) studied wastewater reuse to irrigate lettuce. They reported that more than 80% of nitrogen and 77% of phosphorous were removed from the settled primary sewage by applying it to the roots of lettuce. It is possible to irrigate vegetables with wastewater where a large quantity of nutrients is efficiently removed by vegetable. High nutritive value in wastewater reduces fertilizer application rates and increases productivity of poor fertility soils (Al-Lahham *et al.*, 2003). Kouraa *et al.* (2002) reported that the crop production has a remarkable im-

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provement (about 28%) when irrigating crops either by raw or treated wastewater in comparison to drinking water. In Thailand, Chaimongkol *et al.* (2002) presented that the effluent of sewage treatment plant of Chiang Mai Municipality was suitable for agriculture with the expected yield similar to the one using natural water.

In the South of Thailand, there are many concentrated latex factories. Usually, wastewater from the concentrated latex industry contains high amount of nitrogen originating from natural rubber and ammonia compounds added into the production processes. Most of these factories discharge treated wastewater still containing a high level of nitrogen to the natural river or canals leading to water pollution problems such as algae bloom, dissolved oxygen depletion, etc. Therefore, study of land treatment system was considered and conducted to treat and utilize nitrogen in treated wastewater from the concentrated latex factory

Regarding the latex production process, zinc oxide (ZnO) and sulfuric acid (H_2SO_4) are other two chemicals added into the process. ZnO is added to preserve rubber whereas H_2SO_4 is used in skim rubber production. Therefore, contamination of zinc and sulfate in the concentrated latex wastewater was considered for land treatment system operation.

The main objectives of this study are; 1) to study nitrogen removal efficiency of land treatment system, 2) to study possibility of using the treated wastewater from concentrated latex factory for irrigation and 3) to study mass balance of nitrogen-transformation in land treatment system.

Materials and Methods

Experimental set-up

Pilot-scale experimental land treatment unit was conducted in this research. It consisted of two land treatment units and one control unit (no plantation). The dimension of each unit was 1×2 m (W×L) and 0.2 m soil thickness with 2% slope. Plastic sheet lining was installed at the bottom to prevent infiltration. Each experimental unit was added with sandy clay loam soil. Water convolvulus

and tropical carpet grass were planted into each land treatment unit. Treated wastewater from stabilization pond system of the selected concentrated latex industry in Songkhla was used to irrigate plants and control units with water application rate at 4 cm/week by distribution over each unit twice a day. Each land treatment unit was operated at a different period according to the growing period. The water convolvulus unit was operated for 36 days. Tropical carpet grass and control units were operated for 50 days.

Wastewater analysis

During operation period, influent and effluent were analyzed every 3 days for water convolvulus unit and every 5 days for tropical carpet grass and control unit to determine pH, electrical conductivity (EC), biochemical oxygen demand (BOD_5), nitrite nitrogen (NO_2^--N), nitrate nitrogen (NO_3^--N), ammonia nitrogen (NH_3-N), organic nitrogen, total kjeldahl nitrogen (TKN) and sulfate. Analysis of parameters was done according to the Standard Methods for the Examination of Water and Wastewater (APHA, AWWA and WEF, 1995).

Plant and Soil analysis

Plant and soil samples were analyzed before and after the experimental run. Plants were dried at 65ºC and sent to the Central Analytical Center, Faculty of Natural Resources, Prince of Songkla University, to determine TKN and zinc content. Zinc contents in plants after harvest should less than the maximum safety level of zinc in food (15 mg/100 g wet weight (APFAN, 1994)). Zinc accumulation in plant can be harmful to health. Soil samples were taken from pilot-scale experimental units and sent to the Central Equipment Division, Faculty of Science, Prince of Songkla University, to determine NO_2^--N, NO_3^--N and TKN.

Results and Discussion

Wastewater analysis

Characteristics of wastewater for irrigating experimental land treatment unit are shown in

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Table 1. It demonstrated that pH, sodium adsorption ratio (SAR) and zinc in wastewater were suitable for irrigation. Electrical conductivity (EC) may affect the ability of plants to uptake water but it was at a slight level. Therefore, it was possible to use treated wastewater from the concentrated latex factory for irrigation.

Influent and effluent of land treatment experimental system were analyzed for pH, EC, BOD_5, NO_2^--N, NO_3^--N, NH_3-N, Org-N, TKN and sulfate. The average analysis results are shown in Table 2. The major form of nitrogen in influent was ammonia nitrogen. Plants are able to use this form of nitrogen for growth and it can be accumulated in the soil (Brady and Weil, 1996). Hence, all experimental units provided low level of nitrogen in effluent. In contrast NO_3^--N in effluent of all experimental units were higher than influent. This could occur because of conversion of the NH_3-N to be NO_2^--N and NO_3^--N by nitrifying bacteria under aerobic condition (nitrification process). However, nitrate in the effluent still met the Drinking Water Quality Standard by EPA (US EPA, 1996)

Table 3 shows the average removal efficiencies of TKN, NH_3-N, Org-N, BOD_5 and sulfate for each experimental land treatment unit. It can be clearly observed that tropical carpet grass unit provided the best results. This could occur because of higher density of tropical carpet grass in the land treatment unit. TKN, NH_3-N, Org-N, BOD_5 and sulfate removal efficiencies for water convolvulus and control unit were not significantly different. The results indicated that all experi-

Table 1. Characteristics of treated wastewater from selected concentrated latex factory.

Parameters	Wastewater from selected concentrated latex factory	Degree of restriction on use*		
		None	Slight to moderate	Severe
pH	8.4		Normal range 6.5-8.4	
EC (dS/m)	1.82	< 0.7	0.7-3.0	> 3.0
SAR	1.47	< 3	3-9	> 9
Zn (mg/l)	0.03	-	< 2	-

* FAO (1985)

Table 2. Mean characteristics of influent and effluent of land treatment system.

Parameters	Influent		Effluent		
	Water convolvulus	Carpet grass/Control	Control	Water convolvulus	Carpet grass
TKN (mg/l)	41.00±14.06	33.05±13.79	10.03±4.72	2.47±0.50	8.49±4.16
NH_3-N (mg/l)	35.79±14.15	28.05±13.17	7.16±4.04	0.75±0.65	5.54±3.10
Org -N (mg/l)	5.22±2.23	5.00±2.60	2.88±1.14	1.72±0.67	2.94±1.69
NO_3^--N(mg/l)	0.30±0.12	0.34±0.15	0.64±0.29	0.68±0.39	0.95±0.57
NO_2^--N(mg/l)	0.61±0.28	0.67±0.28	0.26±0.12	0.05±0.06	0.29±0.17
BOD_5 (mg/l)	62±11	65±8	37±7	8±4	45±8
Sulfate (mg/l)	482±243	471±194	308±71	222±62	323±69
pH	8.50±0.52	8.40±0.53	7.90±0.48	7.19±1.38	8.12±0.36
EC (dS/m)	1.88±0.15	1.82±0.15	1.51±0.17	1.10±0.38	1.49±0.28

Note: $\overline{X}$ ± SD

Table 3. Average removal efficiencies of TKN, NH$_3$-N, Org-N, BOD$_5$ and sulfate of land treatment system.

Parameters	Removal efficiency (%)		
	Water convolvulus	Carpet grass	Control
TKN	75±7	92±3	74±6
NH$_3$-N	80±7	97±2	80±5
Organic -N	43±10	61±16	41±12
BOD$_5$	41±6	88±5	31±8
Sulfate	30±15	52±5	28±11

Note: $\overline{X}$ ± SD

Table 4. Nitrogen and zinc contents of water convolvulus after harvest.

Wet weight (kg)	1.572
Dry weight (kg)	0.393
Zinc content (g/kg dry weight)	0.033
Nitrogen content (g/kg dry weight)	22.80
Total nitrogen uptake (g)	8.96

Table 5. Nitrogen and zinc content of carpet grass before and after operation.

	Initial	Final
Wet weight (kg)	2.672	7.700
Dry weight (kg)	0.518	1.702
Zinc content (g/kg dry weight)	0.074	0.024
Nitrogen content (g/kg dry weight)	29.00	14.80
Total nitrogen uptake (g)	15.03	25.18

mental units provided high removal efficiency of TKN and NH$_3$-N. The average removal efficiency for tropical carpet grass, water convolvulus and control unit was found to be 92, 75 and 74% for TKN, and 97, 80 and 80% for NH$_3$-N, respectively.

Plants analysis

After operation for 36 days, water convolvulus was harvested and analyzed for nitrogen and zinc contents. The results are shown in Table 4, showing that zinc accumulation in water convolvulus was 0.033 g/kg dry weight (0.83 mg/100 g wet weight) which was less than the maximum safety level of zinc in food (15 mg/100 g wet weight). Total nitrogen uptake of water convolvulus was 8.96 grams per experimental unit (4.48 g/m^2). The water convolvulus did not achieve mature growth as compared with the natural water convolvulus plant, possibly due to an inadequate trace elements.

Carpet grass was weighed and analyzed for nitrogen and zinc content before and after operation. The results are shown in Table 5, in-

dicating total nitrogen uptake of tropical carpet grass was 10.15 grams per experimental unit (5.10 g/m^2).

Soil analysis

Analysis results of soil before and after experimental run are shown in Table 6. Total nitrogen in initial soil was 272.83 mg/kg dry weight. At the end of experimental run, total nitrogen in soil for tropical carpet grass, water convolvulus and control unit increased to 299.74, 293.26 and 336.95 mg/kg dry weight, respectively. The amount of TKN in soil for water convolvulus and tropical carpet grass units after operation decreased from initial values. This could occur because NH$_3$-N was absorbed by the plant and converted to NO$_3^-$-N by nitrification process that can be observed from the increase in NO$_3^-$-N in final soil of all experimental units.

Nitrogen mass balance

In this research, mass balance was deter-

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Table 6. Analysis results of soil before and after experimental run.

Parameters	Initial	Final		
		Water convolvulus	Tropical carpet grass	Control
TKN (mg/kg)	271.59	245.15	265.38	284.01
NO_3^--N (mg/kg)	1.13	47.49	33.45	52.55
NO_2^--N (mg/kg)	0.11	0.62	0.91	0.40
TN (mg/kg)	272.83	293.26	299.74	336.95

Table 7. Accumulation of nitrogen in land treatment components.

Units	Components				
	Influent (g)[1]	Effluent (g)[2]	Soil (g)	Plant (g)	Volatilized (g)
Water convolvulus	17.20	1.97	4.96	8.96	1.30
Carpet grass	19.41	0.82	6.54	10.16	1.89
Control	19.41	2.92	15.58	-	0.91

[1] = (Mean concentration of N in influent) × (Flow rate of Inf.) × (Operation period)
[2] = (Mean concentration of N in effluent) × (Flow rate of Eff.) × (Operation period)

mined in terms of total nitrogen over operation period to investigate nitrogen transformation and indicate the major removal mechanisms. Total nitrogen accumulation in any components of land treatment system is shown in Table 7.

Mass balances of nitrogen transformations in each experimental unit are presented in Figures 1, 2 and 3. On the mass balance basis, the accumulation of nitrogen in soil for water convolvulus, tropical carpet grass and control units were 28.8, 33.69 and 80.28% of the total inflow, respectively. Nitrogen accumulation in soil for water convolvulus and tropical carpet grass were less than the control unit due to most of nitrogen being taken up by the plants. Nitrogen uptake of water convolvulus and tropical carpet grass were 52.09 and 52.34% of total inflow, respectively. Ammonium (NH_4^+) and nitrate (NO_3^-) are two forms of nitrogen that plants are able to use for growth.

The mass balance of nitrogen indicates that a part of the nitrogen was lost from land treatment. This might occur by ammonia volatilization and denitrification. Due to pH in land treatment ex-perimental units did not favor ammonia volatilization process which occurs at pH 9.2 (Weiner, 2000). Therefore, denitrification was the major mechanism enhancing nitrogen loss in experimental units. Denitrification is the conversion of NO_3^--N to nitrous oxide and nitrogen gas under low oxygen condition. Denitrification occurs when NO_3^--N is present in a soil. Namely, NH_3-N was converted to NO_3^--N under aerobic condition by nitrification process and then NO_3^--N was converted to N_2O and N_2 by denitrification process under low oxygen condition. From the mass balance analysis, nitrogen lost from water convolvulus, tropical carpet grass and control units were 7.59, 9.74 and 4.68% of the total inflow, respectively. Denitrification rate in plant units was higher than in the control unit; this could be possible because of soil moisture of the plant units was higher than of the control unit. High soil moisture content will reduce oxygen level in soil enhancing denitrification rate. Chenfang *et al*. (2000) reported that soil moisture was the influential factor governing nitrogen loss via the process of denitrification.
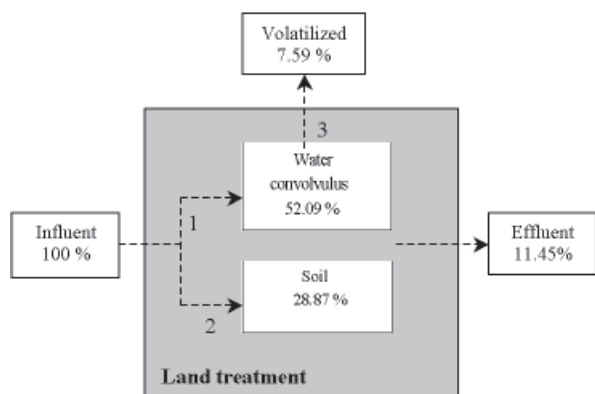
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Figure 1. Nitrogen mass balance of water convolvulus experimental unit.
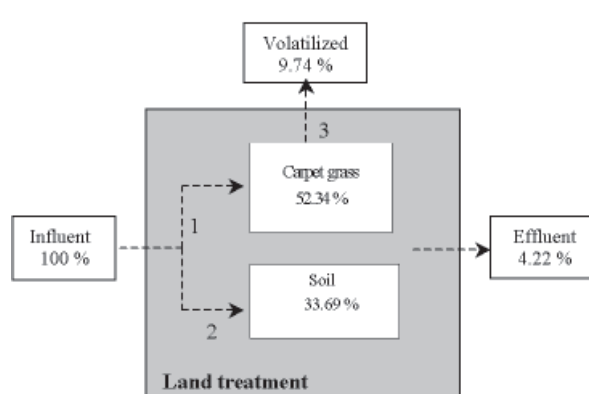**1) plant uptake 2) soil adsorption and 3) nitrification/denitrification**

Figure 2. Nitrogen mass balance of tropical carpet grass experimental unit.
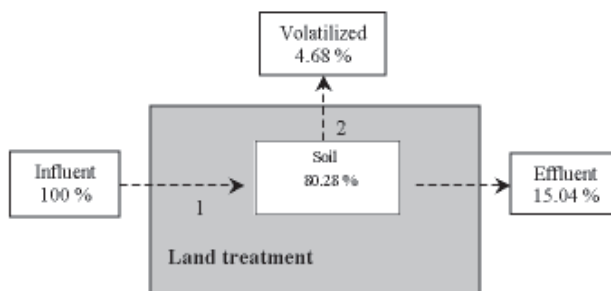**1) plant uptake 2) soil adsorption and 3) nitrification/denitrification**

Figure 3. Nitrogen mass balance of control experimental unit.
1) soil adsorption, 2) nitrification/denitrification

Annette and Ralf (1998) reported that N_2O and N_2 release by denitrification increased with increasing soil moisture content.

Conclusion

Land treatment system provided a high nitrogen removal efficiency. In the control unit, the major nitrogen removal mechanism was soil accumulation. Nitrogen accumulation in soil for water convolvulus and tropical carpet grass were less than in the control unit due to most of accumulated nitrogen in soil being taken up by plants. Plant uptake was the major nitrogen removal mechanism for the water convolvulus and tropical carpet grass units. Tropical carpet grass provided the highest nitrogen removal efficiency. It indicates that the higher density of plants in the land treatment system enhances nitrogen removal efficiency.

Water convolvulus did not achieve mature growth as compared with the natural water convolvulus plant, possibly due to inadequate trace elements. However, it is possible to irrigate vegetables with the treated wastewater with the addition of chemical fertilizer or manure to yield a commercial crop.

Acknowledgments

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