

# DIVISION S-10—WETLAND SOILS

## Nitrogen Distribution in Soils of Constructed Wetlands Treating Lagoon Wastewater

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### ABSTRACT

Constructed wetlands have the potential to be used for treatment of N-rich livestock wastewater. Our objectives were to evaluate both the time effect and increasing N loading rates on soil N distribution and  $\text{NH}_4^+\text{-N}$  concentration in surface-pore water of constructed wetlands. A 5-yr study in North Carolina investigated two wetland systems that treated swine lagoon wastewater. Wetland System 1 was planted to a *Schoenoplectus americanus* (Pers.) Volkart ex Schinz & R. Keller, *S. tabernaemontani* (K.C. Gmel.) Palla, *Scirpus cyperinus* (L.) Kunth, and *Juncus effusus* L. plant community, and Wetland System 2 was planted to a *Typha angustifolia* L., *T. latifolia* L., and *Sparganium americanum* Nutt. plant community. Nitrogen loading rates were increased annually from 0.6 to 2.7  $\text{g m}^{-2} \text{d}^{-1}$ . Soils were analyzed for total N annually. Surface-pore water was sampled with equilibrators and analyzed for  $\text{NH}_4^+\text{-N}$ . Although the total N accumulation significantly increased with time in both systems, total soil N accumulation by depth did not differ significantly between systems. Distribution profiles in the surface-pore water column showed that  $\text{NH}_4^+\text{-N}$  was transported upward into surface water at N loading rates from 1.2 to 2.7  $\text{g m}^{-2} \text{d}^{-1}$ . As total N loading rates increased annually in both wetland systems, soil pore water had higher levels of  $\text{NH}_4^+\text{-N}$  but N removal efficiency of the wetlands sharply decreased. Accumulation of high levels of  $\text{NH}_4^+\text{-N}$  ( $>200 \text{ mg L}^{-1}$ ) in soil pore water could negatively affect long-term ability of wetland systems to treat wastewater with high N levels.

MANAGEMENT OF SWINE WASTE is a national concern since the traditional treatment method of anaerobic lagoon-spray field is effective only when large tracts of cropland are available and neighbors are some distance from application areas. When land and demographic conditions are limiting, other waste management systems are needed that will reduce the contamination hazard of water resources by concentrated livestock wastewater.

Constructed wetlands, as a component of an on-farm total waste management system, are less land intensive than the traditional lagoon-spray field system (Cronk, 1996; Humenik et al., 1999; Knight et al., 2000; Hunt and Poach, 2001). They have been used for many years in wastewater treatment, and a significant understanding exists on the role of plants, soil, water, and microbial processes that affect nutrient removal from wastewater in municipal wetland treatment systems (Gersberg et

al., 1986; Hammer, 1989; Kadlec and Knight, 1996). When N-rich wastewater is applied to constructed wetlands, the major expected removal mechanism of N is nitrification-denitrification and to a lesser extent, plant uptake and ammonia volatilization (Watson et al., 1989). However, the magnitude of these mechanisms is affected by wetland operational parameters such as water depth, retention time, N form and loads, and environmental conditions such as temperature and availability of dissolved oxygen.

In a nationwide survey study, Knight et al. (2000) found that wetland systems constructed for livestock wastewater treatment had an average water depth of 38 cm and  $\text{NH}_4^+\text{-N}$  was the prevalent form of N with average inflow concentrations of 366  $\text{mg L}^{-1}$ . High  $\text{NH}_4^+\text{-N}$  concentrations and water depth ( $>10 \text{ cm}$ ) can adversely affect biological N removal by nitrification-denitrification (Reed and Brown, 1992). Moreover, high  $\text{NH}_4^+\text{-N}$  concentrations ( $>200 \text{ mg L}^{-1}$ ) can negatively affect plant growth and effectiveness of wetlands built to treat ammonium-rich waters (Clark and Baldwin, 2002). Research on constructed wetlands for dairy wastewater treatment reported by Majer Newman et al. (2000) indicated that N was removed mainly by sedimentation but very little by denitrification, possibly because of N overloading and ammonia accumulation. Another study with wetlands treating dairy wastewater reported that plant uptake and soil accumulation were considered the major mechanisms to reduce N at water depths of 15 to 30 cm (Shamir et al., 2001). However, Hunt et al. (2002, 2003) concluded that denitrification was likely a major mechanism to reduce N in constructed wetlands operated at shallow water depth ( $<10 \text{ cm}$ ) that treated swine lagoon wastewater. These divergent conclusions about N retention in constructed wetlands treating livestock wastewater indicate that there is a need for better understanding of component functions for N recycling in constructed wetlands. In this respect, the role of the soil substrate is important to the overall constructed wetland function of enhancing water quality since soil is the supporting medium for vegetation, habitat for microbes involved in N cycling, and transitional storage of organic and inorganic N (Good and Patrick, 1987; Reddy et al., 1989a, b).

The wetlands of our study were investigated from 1993 through 1997 for their effectiveness to treat swine lagoon wastewater subject to shallow water depth ( $<10 \text{ cm}$ ) and a wide range of N loading rates (Hunt et al., 2002). Since the N loading rates were increased annu-

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Published in Soil Sci. Soc. Am. J. 67:1943–1951 (2003).

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**Abbreviations:** NRCS, Natural Resource Conservation Service; ORP, oxido-reduction potential; TKN, total Kjeldahl N.

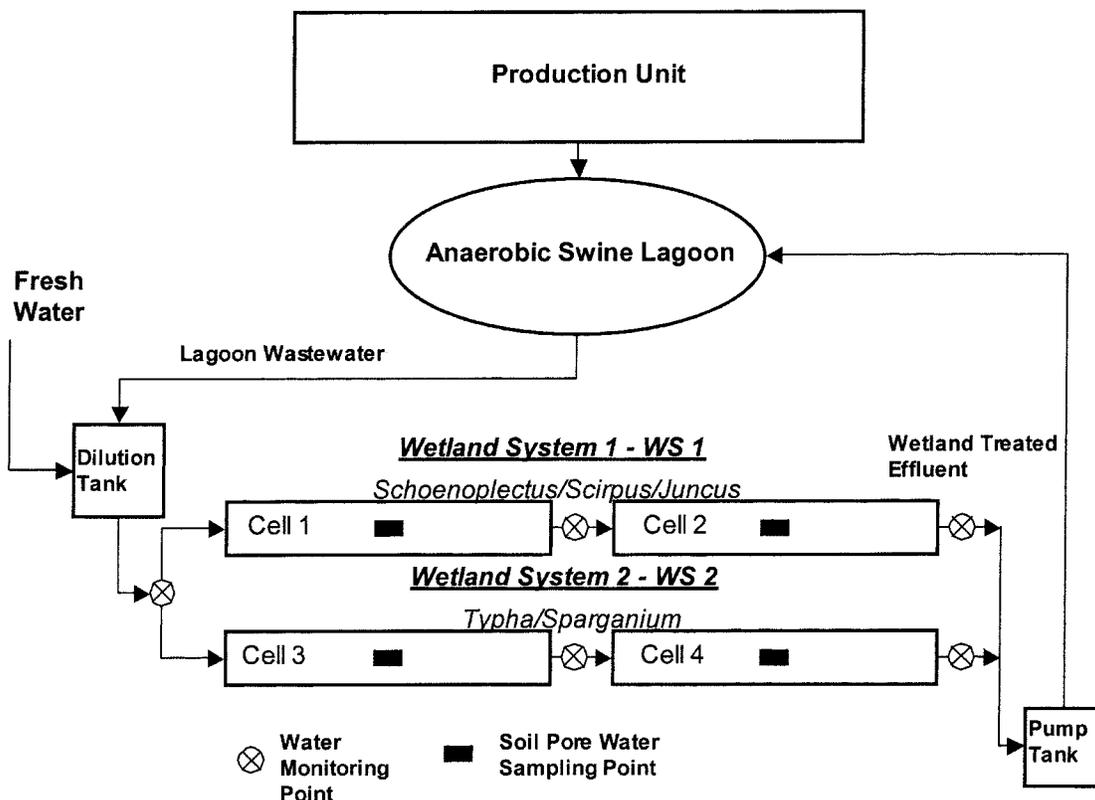


Fig. 1. Schematic of constructed wetlands.

ally, it was expected that the resulting N accumulation in wetland soils would increase with time, as well as the N contribution of plant litter to total soil N storage. The objectives of this study were (i) to evaluate the time effect (5 yr) on storage and distribution of total N in wetland soil components and (ii) to assess the effect of N loading rates on  $\text{NH}_4^+\text{-N}$  concentration distribution in surface-pore water columns of two constructed wetland systems treating swine lagoon wastewater.

## MATERIALS AND METHODS

### Study Site

In 1992, two pilot surface-flow wetland systems for treatment of swine lagoon wastewater were constructed in Duplin Co., NC. The Natural Resource Conservation Service (NRCS) designed the systems to treat wastewater supplied by an adjacent anaerobic lagoon (NRCS, 1991). Constructed wetlands consisted of four  $3.6 \times 33.5\text{-m}$  wetland cells arranged in two parallel sets of two end-to-end connected cells (Fig. 1). Lengthwise slope of wetland cells was 0.2%. Soil at the site where the wetland cells were excavated was a Bonneau loamy sand (loamy, siliceous, subactive, thermic Arenic Paleudults). Once wetland cells were excavated, a 0.3-m compacted clay liner for seepage control was installed. Topsoil was backfilled to cover the clay liner, creating a 0.25-m soil layer above the liner (Table 1).

Four cells were manually planted to native wetland vegetation in May 1992 with nursery-grown, vegetative propagules. A set of two connected cells was planted to a plant community that contained three species of bulrushes (*Schoenoplectus americanus*, *S. tabernaemontani*, and *Scirpus cyperinus*) and rush (*Juncus effusus*); they will hereafter be referred to as

*Wetland System 1*, which contains Cells 1 and 2. A second set of two cells was planted to a plant community that contained two species of cattails (*Typha angustifolia* and *T. latifolia*) and bur-reed (*Sparganium americanum*); they will hereafter be referred to as *Wetland System 2*, which contains Cells 3 and 4. These two wetland systems were flooded with fresh water and kept in shallow water conditions ( $<0.15\text{ m}$ ) until the start of wastewater application in June 1993.

### Wastewater Treatment

Wastewater flow and chemical properties were monitored with flow meters and automated samplers, respectively, at the inlet and outlet of each wetland system as described by Hunt et al. (2002). Anaerobic lagoon liquid was pumped to a tank and diluted with fresh water to adjust nutrient loading rates before wetland treatment (Fig. 1). Loading rates were obtained by adjusting the fresh water to wastewater flow ratio into the dilution tank (Table 2). Stone et al. (2002) reported details on design and performance of these same wetlands. Much of the total N occurred predominantly in the inorganic form in both wetland inflow and outflow. Ammonium-N accounted for  $\approx 95\%$  of the total N. Mean inflow  $\text{NH}_4^+\text{-N}$  concentrations were initially  $35\text{ mg L}^{-1}$  in 1993 and increased with increased loading rates to  $225\text{ mg L}^{-1}$  in 1997. Mean outflow  $\text{NH}_4^+\text{-N}$  concentrations were initially  $2\text{ mg L}^{-1}$  in 1993 and increased to  $58\text{ mg L}^{-1}$  at the higher loading rates in 1997. At lower loading rates ( $<0.9\text{ g m}^{-2}\text{ d}^{-1}$ ), the  $\text{NH}_4^+\text{-N}$  concentration reduction efficiencies were 92 to 95%. However, concentration reduction efficiencies declined with increasing loading rates. Concentration reduction efficiencies for  $\text{NH}_4^+\text{-N}$  at the higher loading rates ( $2.5\text{--}2.7\text{ g m}^{-2}\text{ d}^{-1}$ ) were  $\approx 75\%$ . Mean pH values were 7.8 to 8.3 in the inflow and 7.7 to 8.0 in the outflow. During the study period, water was maintained at

**Table 1. Surface soil (0–20 cm) properties in the constructed wetland cells before flooding in 1992.**

Property	Mean	SD	n
Texture, g kg <sup>-1</sup>			
Sand	863	12	4
Silt	101	5	4
Clay	36	5	4
pH in water	5.4	0.2	4
Organic C, mg kg <sup>-1</sup>	3.6	0.3	4
Total N, mg kg <sup>-1</sup>	150	7	4

shallow depths (<10 cm), and the systems had a mean residence time of 12 to 14 d.

### Litter and Soil Sampling and Analysis

Plant litter material, which consisted of dead shoots above the soil surface, was collected each year when vegetation reached peak growth in three 0.25-m<sup>2</sup> plots selected at random within the cell. Plant material samples were oven dried at 65°C to constant moisture, weighed, and ground. Samples were then digested with a block digester following procedures described by Gallaher et al. (1976) and analyzed for total Kjeldahl N (TKN). Soil cores (2.2 by 20 cm; n = 8 subsamples) were obtained from each wetland cell at three sites (upper end, middle, and lower end). Soil cores were sectioned by 5-cm increments down to 20 cm, and subsamples combined into one composite sample per depth and per site. Each year, this sampling procedure provided a total of 12 samples (three sites at four depths) per cell. Soil samples did not include clay material from the bottom liner. Soil samples were transported on ice to the laboratory. Soil pH was measured in wet samples (1:1 soil to water mixture) with a combination electrode. Soil samples were air dried, crushed, passed through a 2-mm sieve, digested, and analyzed for TKN according to Gallaher et al. (1976). Extractable NH<sub>4</sub><sup>+</sup>-N and NO<sub>3</sub><sup>-</sup>-N were determined according to Keeney and Nelson (1982). Total Kjeldahl N in plant and soil digestates, and NH<sub>4</sub><sup>+</sup>-N and NO<sub>3</sub><sup>-</sup>-N in soil extracts were determined by automated analysis with a Technicon Auto Analyzer II (Technicon Instruments Corp., Tarrytown, NY). Total N was the sum of TKN plus NO<sub>3</sub><sup>-</sup>-N. Soil texture was determined by the micropipette method (Miller and Miller, 1987), and bulk density was determined by the core method (Blake and Hartge, 1986). Soil organic carbon was determined by dry combustion with a LECO C-analyzer (Leco Corp., St. Joseph, MI).<sup>1</sup>

### Water–Soil Column Sampling and Analysis

Plexiglas soil pore water equilibrators were used once a year (August–September) to sample interstitial soil water and the overlying water column from 1994 to 1997. Each equilibrator had two parallel sets of 3-mL compartments spaced at 1-cm intervals, with a total of 23 compartments per equilibrator (Simon et al., 1985). Both sides of the equilibrator were covered with rectangular 0.2- $\mu$ m polycarbonate membranes (Nucleopore Corp., Pleasanton, CA) and sealed with plexiglas covers. One side was assembled first with the membrane and plexiglas cover to hold water. When each compartment was filled with distilled-deionized water, the equilibrator was sealed with the other membrane and cover. Equilibrators were stored in plastic containers filled with distilled-deionized water and bubbled with N<sub>2</sub> gas for 24 h. After the N<sub>2</sub>-bubbling

<sup>1</sup>Mention of trademark, proprietary product, or vendor does not constitute a guarantee or warranty of the product by the U.S. Department of Agriculture and does not imply its approval to the exclusion of other products or vendors that also may be suitable.

**Table 2. Mean daily N total loading rates, and cumulative annual total N mass load applied to constructed Wetland System 1 (WS1; *Schoenoplectus/Scirpus/Juncus*) and Wetland System 2 (WS2; *TyphalSparganium*).**

Year	Mass Load <sup>†</sup>		Cumulative Mass Load	
	WS1	WS2	WS1	WS2
	g m <sup>-2</sup> d <sup>-1</sup>		g m <sup>-2</sup>	
1993	0.5	0.6	104	122
1994	0.6	0.6	310	354
1995	0.8	0.9	611	685
1996	1.2	1.8	1043	1326
1997	2.7	1.5	1914	1803

<sup>†</sup> Adapted from Hunt et al. (2002).

period, plastic containers were covered with a lid and sealed. This maintained the anoxic condition of the equilibrators during transport. Equilibrators were installed in both Wetland System 1 and Wetland System 2 at approximately the center of each cell (Fig. 1). When the equilibrators were inserted in the soil, the top two compartments were in the water column, above the soil–water interface, in 1994, 1995, and 1996. In 1997, the water column was sampled more extensively, and, except for the equilibrator in Cell 2, which was left with two compartments above the soil–water interface, equilibrators were installed with eight compartments left in the water column. Equilibrators remained in the field for 14 d to reach equilibrium under continuous flooding conditions; average daily water temperature varied from 23 to 26°C. Immediately after the equilibrators were taken out of the constructed wetland, the compartments were sampled with a syringe. Samples were placed in 4-mL plastic vials, acidified (1  $\mu$ L of 50% H<sub>2</sub>SO<sub>4</sub>) to pH 2, and transported on ice to the laboratory. Ammonium-N and NO<sub>3</sub><sup>-</sup>-N were analyzed with a Technicon Auto Analyzer II with USEPA Methods 350.1 and 363.2 (USEPA, 1983).

The NH<sub>4</sub><sup>+</sup>-N water profiles were used to estimate steady state diffusive flux according to Fick's law (Lerman, 1988):

$$J_i = -\Phi^2 D_i dc/dz,$$

where  $J_i$  is the flux of the dissolved species  $i$  per unit area and time;  $\Phi$  is the porosity of the soil;  $D_i$  is the diffusion coefficient of species  $i$ ; and  $dc/dz$  is the concentration gradient with depth. Concentration gradients between surface and soil pore water were estimated by linear regression between depths of -4 to +2 cm for all cells. Porosities near the water–soil interface were assumed to be 0.86 (*Schoenoplectus tabernaemontani*) for Wetland System 1 and 0.90 (*Typha* sp.) for Wetland System 2 according to Watson and Hobson (1989). A diffusion coefficient of  $19.8 \times 10^{-6}$  cm<sup>2</sup> s<sup>-1</sup> at 25°C was used for NH<sub>4</sub><sup>+</sup>, according to Li and Gregory (1974).

### Statistical Analysis

Our study was performed on a large-scale system that lacked replication. Treatment replication was not possible because of the great cost of wetland cell construction and monitoring equipment. However, one viable statistical method to remedy the lack of replication is repeated measures design analysis. With such analysis, one could answer the question whether the time trajectories in two systems start to diverge from the onset of the experiment (Oksanen, 2001). In our study, we used repeated measures designs with subsampling and different time sampling variation error effects (Green, 1993). Analysis of variance for a single-factor design with repeated measures was used to test the hypothesis of no treatment effect N accumulated in litter. An ANOVA for repeated measures split-split block design (system  $\times$  cell  $\times$  depth  $\times$

year) was used to test the hypotheses that there were not differences in N accumulation between treatment systems or soil depths across time; the design included the test of the following interactions effect: system  $\times$  depth, cell  $\times$  depth (system), and cell (system). For multiple comparisons among means we used the LSD test (Schlotzhauer and Littell, 1987). Data were analyzed by means and SDs (proc MEANS), analysis of variance (proc ANOVA), and regression (proc REG) with SAS Version 8 (SAS Institute, 1999).

## RESULTS AND DISCUSSION

### Litter and Biomass Nitrogen Storage

Plant litter accumulation is a major precursor of soil development and nutrient storage in wetlands, although it is not considered part of wetland soil (Kadlec and Knight, 1996). Litter production depends on aboveground (shoots) biomass. In our wetlands, total N stored in aboveground live plant biomass had mean annual values of 35.4 g m<sup>-2</sup> yr<sup>-1</sup> in Wetland System 1 and 31.7 g m<sup>-2</sup> yr<sup>-1</sup> in Wetland System 2 (Hunt et al., 2002). In addition, total N stored in belowground biomass (roots and rhizomes) had values of 6.0 g m<sup>-2</sup> yr<sup>-1</sup> in Wetland System 1 and 4.8 g m<sup>-2</sup> yr<sup>-1</sup> in Wetland System 2 (1993 and 1994, unpublished data). In 5 yr, residues from litter and root decomposition contributed to the development of a 2-cm layer of muck over the mineral soil. During the 5 yr of this investigation, there was a wide range of annual dry matter litter production, 351 to 744 g m<sup>-2</sup> (Table 3). Significant differences in dry matter production and N storage in litter between Wetland System 1 and Wetland System 2 occurred only in 1993. Total N stored in plant litter ranged from 4.5 to 12 g m<sup>-2</sup>. Causes of wide ranges for dry matter accumulation and N storage in litter were related to yearly variable plant growth and dry matter production, community composition, and insect–plant disease pressure (Hunt et al., 2002). However, the 5-yr total N means were very similar for Wetland System 1 and Wetland System 2, 8.3 and 7.0 g m<sup>-2</sup>, respectively. These values are within typical total N content range of 2 to 20 g m<sup>-2</sup> in litter of constructed wetlands (Kadlec and Knight, 1996). Estimated mean annual total N storage in plant biomass (standing crop plus litter) was 43.7 g m<sup>-2</sup> yr<sup>-1</sup> for Wetland System 1 and 38.7 g m<sup>-2</sup> yr<sup>-1</sup> for Wetland System 2. However, at the time of vegetation sampling, total N stored in litter represented a small portion (12–14%) relative to the total N assimilated in plant biomass. Since the storage

in plant biomass and litter change with seasonal plant growth and death rates, decomposition processes would release most of the stored N to the soil.

### Soil Nitrogen Storage

Under wetland conditions, there is a net accretion of total soil N because of accumulation of soil organic matter. Maximum accumulation of total soil N in constructed wetlands ranges from 100 to 1000 g m<sup>-2</sup> (Faulkner and Richardson, 1989). Extractable N, which represented the easily exchangeable NH<sub>4</sub><sup>+</sup>-N and soluble NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup>-N, was always <10% of the total soil N stored in both wetland systems. At the highest N loading rate in 1997, soil extractable NH<sub>4</sub><sup>+</sup>-N was 70 mg kg<sup>-1</sup> in Wetland System 1 and 80 g kg<sup>-1</sup> in Wetland System 2. With wetland cell soil bulk densities (1.42–1.54 g cm<sup>-3</sup>) we estimated a mean soil extractable NH<sub>4</sub><sup>+</sup>-N content of 5 and 6 g m<sup>-2</sup> in Wetland System 1 and Wetland System 2, respectively. Differences in mean extractable NH<sub>4</sub><sup>+</sup>-N between the two systems were not significant ( $P < 0.10$ ). Extractable soil NO<sub>3</sub><sup>-</sup>-N was highly variable and detected only in the second cells of each system. Detection of NO<sub>3</sub><sup>-</sup>-N was related to aerobic episodes because of draining of the wetland cells, and at much lower levels than extractable NH<sub>4</sub><sup>+</sup>-N (11 to 40 g kg<sup>-1</sup>).

The time effect on N accumulation and distribution was discerned by testing the following three null hypotheses ( $P < 0.10$ ): (i) no time (year) effect on total soil N storage; (ii) no difference in total soil N storage between systems; and (iii) no differences among depths. The ANOVA (split-split block design) analysis indicated that there was a significant effect of time on total N storage but no significant difference between systems or the interaction system  $\times$  depth. Difference in total N accumulation by depth was significant but not the interactions cell  $\times$  depth (system), and cell (system). The time (year) effect on total N concentration at each soil depth in each cell and system is shown in Fig. 2. Lowest total soil N content occurred in 1993 because newly planted and flooded constructed wetlands produced very little litter in 1992. In subsequent years, litter and belowground plant material accumulation and decomposition supported buildup of total soil N. With the exception of Cell 1 in 1995, in all four cells, soil N content was significantly higher in the upper-profile 5-cm layer than in the lower-profile layers (5–10, 10–15, and 15–20 cm). With respect to the initial N content in 1992 (46 g m<sup>-2</sup>), the upper 5-cm layer stored an additional 36 g m<sup>-2</sup> in Wetland System 1 and 34 g m<sup>-2</sup> in Wetland System 2 in the 5-yr study. On average for both systems, the upper 5-cm layer stored 34% of the total soil N. The 5-yr mean total soil N concentration decreased logarithmically with depth (D) in both wetlands: Wetland System 1 [N mg kg<sup>-2</sup> = 578 - 101ln (D cm);  $r^2 = 0.95$ ;  $n = 4$ ]; and Wetland System 2 [N mg kg<sup>-2</sup> = 553 - 97 Ln (D cm);  $r^2 = 0.97$ ;  $n = 4$ ]. When total soil N data were pooled by wetland system, mean total N concentrations were not significantly different between Wetland System 1 and Wetland System 2 within any year ( $P < 0.10$ ). We concluded from these

**Table 3. Dry matter and total N accumulation in litter of constructed Wetland System 1 (WS1; *Schoenoplectus/Scirpus/Juncus*) and Wetland System 2 (WS2; *Typha/Sparganium*).**

Year	Dry matter			Nitrogen		
	WS1	WS2	LSD0.10	WS1	WS2	LSD0.10
	— g m <sup>-2</sup> —			— g m <sup>-2</sup> —		
1993	662	415	†	12.0	4.5	†
1994	744	669	ns‡	9.7	6.1	ns
1995	450	514	ns	5.9	8.8	ns
1996	351	476	ns	6.2	8.2	ns
1997	421	420	ns	7.7	7.6	ns
Mean	525	499	ns	8.3	7.0	ns

† LSD significant ( $P < 0.10$ ),  $n = 6$ .

‡ ns, nonsignificant.

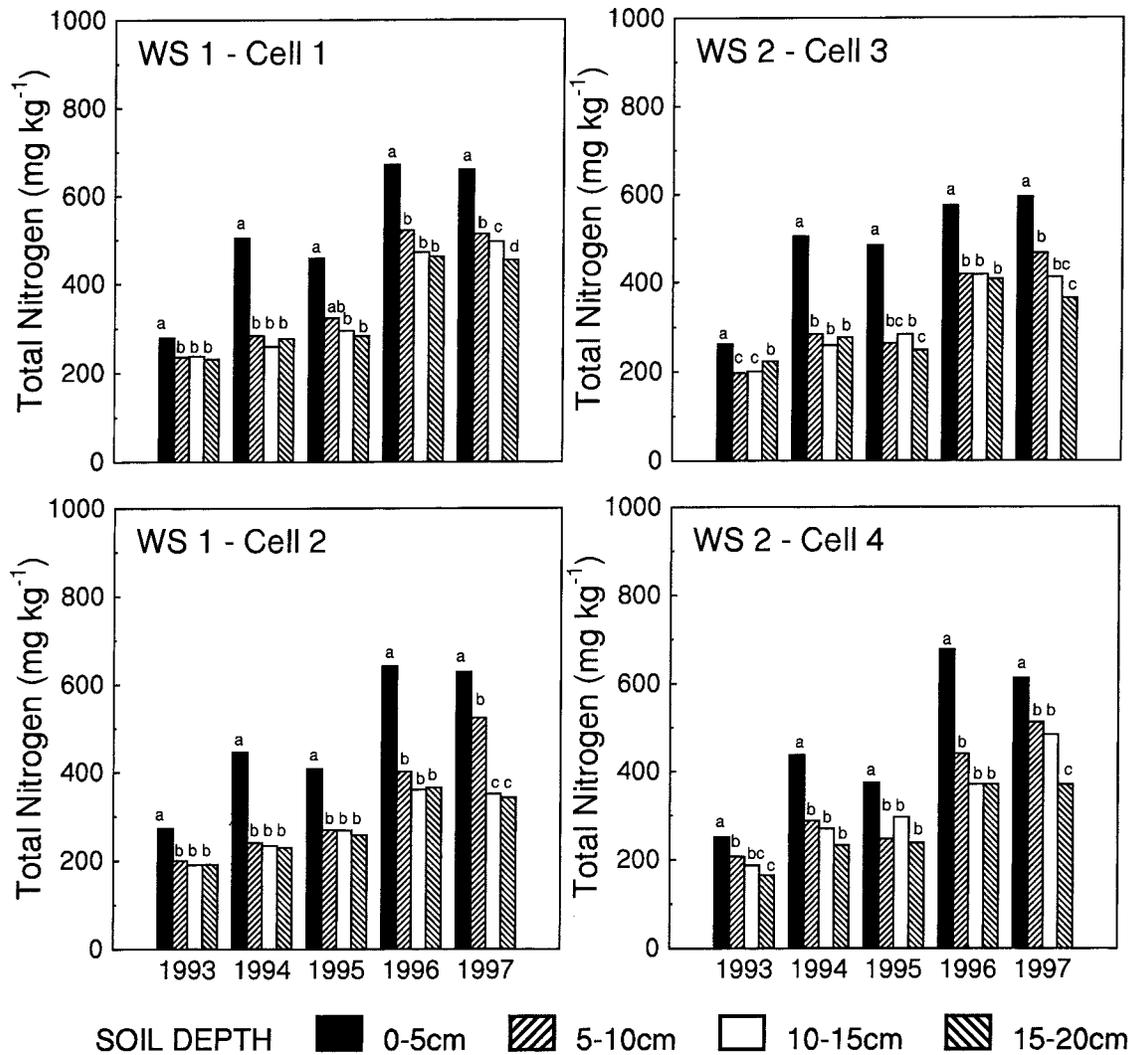


Fig. 2. Mean annual soil N concentrations by depth in Cells 1 and 2 of Wetland System 1 (WS1, *Schoenoplectus/Scirpus/Juncus*) and in Cells 3 and 4 of Wetland System 2 (WS2, *Typha/Sparganium*). Means for each soil depth followed by the same letter are not significantly different ( $P < 0.10$ ) within the same year.

results that there was no difference in the accumulation pattern of soil N by depth in both wetland systems. In addition, the 5-yr net N accumulation, with respect to the initial N content in 1992, was not significantly different between Wetland System 1 ( $106 \text{ g m}^{-2}$ ) and Wetland System 2 ( $100 \text{ g m}^{-2}$ ). Yet, this accumulation in our study was a small (<6%) portion of the total N applied ( $1800 \text{ g m}^{-2}$ ) during the study period. Lack of differences between stored total N in both Wetland Systems 1 and 2 and the small accumulation with respect to total N applied was probably because of internal recycling. According to Mitsch and Gosselink (1993), this is a strategy common for natural wetland ecosystem development; most of the plant N demand is supplied by internal recycling in wetlands with little response to N addition from an external source. Most probably, N uptake reached a maximum in both systems, beyond which, addition of N with wastewater did not greatly increase plant and/or litter production.

## Surface-Soil Pore Water

### Distribution Profiles

Detailed profiles show that at low N loading rates,  $\text{NH}_4^+\text{-N}$  concentrations decreased with depth for all cells in 1994 (Fig. 3). A significant decrease in  $\text{NH}_4^+\text{-N}$  concentration below the depth of  $-2 \text{ cm}$  indicated that  $\text{NH}_4^+\text{-N}$  likely was transported from the surface water downward into soil pore water. Concentration profiles for Cells 1, 2, and 3 in 1995 and 1996 and for all cells in 1997 were different from 1994 profiles. They showed development of a peak below the water-soil interface. In 1997, the  $\text{NH}_4^+\text{-N}$  concentrations increased pronouncedly ( $100$  to  $300 \text{ mg L}^{-1}$ ) in all cells from the soil-water interface down to a depth of  $\approx 3$  to  $5 \text{ cm}$ , from where  $\text{NH}_4^+\text{-N}$  decreased further down with soil depth. Lower  $\text{NH}_4^+\text{-N}$  concentrations above and below the peak indicated that  $\text{NH}_4^+\text{-N}$  was being transported upward and downward within the pore water column;  $\text{NH}_4^+\text{-N}$  concentration maximum suggests that a combi-

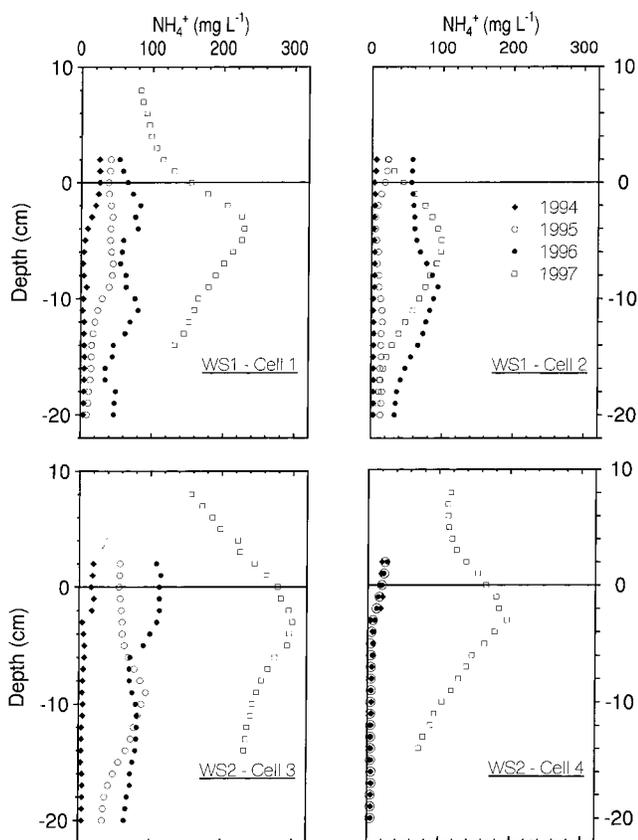


Fig. 3. Surface water and soil pore water profiles of ammonium-N concentration with depth (1-cm interval) in Wetland System 1 (WS1, Cells 1 and 2, *Schoenoplectus/Scirpus/Juncus*) and Wetland System 2 (WS2, Cells 3 and 4, *Typha/Sparganium*). Solid horizontal lines at depth = 0 indicate the soil–water interface boundary. Each data point is the mean of duplicate samples.

nation of simultaneous processes such as plant uptake, soil adsorption, microbial assimilation, and organic matter mineralization was probably responsible for the peaks.

Distribution of surface–soil pore water  $\text{NH}_4^+$ -N concentration had different profiles with depth in each cell

as the wetland systems aged and received increasing N loading rates. Profiles in Fig. 3 show the wetland treatment attenuation of  $\text{NH}_4^+$ -N along the wetlands. Ammonium-N concentration levels were consistently low in surface and pore water of the second cells (2 and 4) than in the first cells (1 and 3) of each system. Nitrates in soil pore water were always very low ( $<0.6 \text{ mg L}^{-1}$ ). Significant levels of nitrates were occasionally found in water column and pore water samples at shallow soil depths (0 to  $-2 \text{ cm}$ ) only when wetland cells were drained 1 or 2 d because of application malfunction or maintenance work (data not presented). When this occurred in any of the cells, the four cells were sampled again to obtain samples from 14-d continuous flooding period.

### Diffusive Gradients and Fluxes

Disappearance of  $\text{NH}_4^+$ -N in free water surface wetland treatment is attributed to combination of a large number of processes that transport N compounds from one point to another in wetlands (sedimentation, diffusion, litter fall, plant uptake, ammonia volatilization, and sorption) and molecular N transformation processes (mineralization, nitrification-denitrification, fixation, and assimilation) (Kadlec and Knight, 1996). Although fluxes explained a small portion (5–12%) of the material transport required to remove  $\text{NH}_4^+$ -N in the wetlands, they were useful to discern the possible mechanisms for N removal.

In contrast with 1994, all  $\text{NH}_4^+$ -N fluxes in 1997 were positive, indicating that  $\text{NH}_4^+$ -N was transported from the soil pore water into surface water. Upward fluxes indicated that  $\text{NH}_4^+$ -N was being removed above the surface–pore water interface, probably because of gaseous losses.

A positive concentration gradient indicated that  $\text{NH}_4^+$ -N was transported from the water surface into soil. Such transport, with negative fluxes, across the water–soil interface was prevalent at N loading rates  $<0.6 \text{ g m}^{-2} \text{ d}^{-1}$ . Negative fluxes were present for all cells in 1994, for Cells 2 and 4 in 1995, and Cell 4 in 1996 (Table 4). These results are compatible with the

Table 4. Mean concentration gradients and  $\text{NH}_4^+$ -N fluxes at the soil–water interface in constructed Wetland System 1 (*Schoenoplectus/Scirpus/Juncus*) and Wetland System 2 (*Typha/Sparganium*).

Year	Wetland System	N applied <sup>†</sup>	Cell	Gradient $dc/dz$ <sup>‡</sup>	$r^2$	Diffusive flux
		$\text{g m}^{-2} \text{ d}^{-1}$		$\text{mg L}^{-1} \text{ cm}^{-1}$		$\text{mg m}^{-2} \text{ d}^{-1}$
1994	1	0.6	1	2.8	0.82	-41
			2	0.4	0.68	-6
	2	0.6	3	2.4	0.61	-37
			4	2.6	0.72	-40
1995	1	0.8	1	-0.4	ns§	ns
			2	3.4	0.96	-50
	2	0.9	3	-0.76	0.76	+12
			4	3.3	0.98	-51
1996	1	1.2	1	-4.6	0.83	+68
			2	-0.7	0.76	+10
	2	1.8	3	1.3	NS§	NS
			4	3.0	0.92	-46
1997	1	2.7	1	-20.9	0.98	+322
			2	-12.5	0.99	+184
			3	-8.5	0.88	+131
			4	-7.9	0.75	+122

<sup>†</sup> N application rate estimated at the inlet of each wetland system.

<sup>‡</sup> Diffusion gradients estimated between  $+2\text{-cm}$  (above soil surface) and  $-4\text{-cm}$  depth from concentration profiles.

§ NS, indicates gradient had nonsignificant slope;  $n = 7$  ( $P < 0.05$ ).

observation that plant uptake was probably a major mechanism for N removal at the lowest loading rates (Hunt et al., 2002). Nonsignificant fluxes for Cell 1 in 1995 and Cell 3 in 1996 indicated a uniform distribution of  $\text{NH}_4^+\text{-N}$  across the surface-pore water interface.

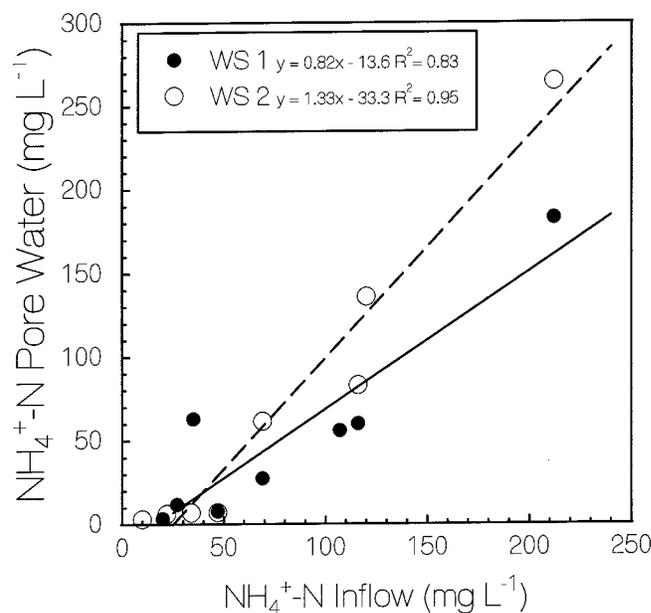
Significant ammonia volatilization is promoted by soil and water alkaline conditions ( $\text{pH} > 8.5$ ) and environmental conditions (temperature and wind) (Reddy and Patrick, 1984). In this study, the acid soil pH changed to neutral values as a result of both flooding and the additional alkalinity of the wastewater (Table 5). However, under similar water temperature, pH, and N loads as of 1997, Poach et al. (2002) found that ammonia losses with respect to the total N load were 14% in Wetland System 1 (bulrush) and 16% in Wetland System 2 (cattails). They concluded that ammonia volatilization was not responsible for removing the majority of N from swine wastewater in these constructed wetlands.

Reddy et al. (1976) attributed upward diffusion and subsequent oxidation of  $\text{NH}_4^+\text{-N}$  as a significant removal mechanism of  $\text{NH}_4^+\text{-N}$  applied to a flooded soil. In flooded soils, both nitrification and denitrification can proceed at the same time, and  $\text{NH}_4^+\text{-N}$  levels are greatly influenced by the presence of a thin, surface-oxidized aerobic layer and anaerobic soil layers. Ammonium-N in the oxidized layer is readily oxidized to  $\text{NO}_3^-\text{-N}$  by microbes. In turn,  $\text{NO}_3^-\text{-N}$  diffuses down into anaerobic layers and is depleted by microbial denitrification (Reddy and Patrick, 1984). Evidence of nitrification-denitrification in these wetlands was found by denitrification enzyme assays and soil oxido-reduction potential (ORP) measurements (Hunt et al., 2002, 2003). Soil ORP measurements at the 2-cm depth indicated reduced soil conditions within the  $\text{NO}_3^-\text{-N}$  reduction range in Wetland System 1 (+130 to +308 mV) and in Wetland System 2 (+105 to +196 mV). Nitrate-N is unstable and denitrified under anaerobic conditions with ORP values of 220 mV in near-neutral soils (Gambrell and Patrick, 1978). Higher ORP readings in Wetland System 1 indicated that higher oxygen content was likely available for nitrification. In turn, denitrification rates estimated

**Table 5. Flooded soil pH profiles with depth (5-cm intervals) in constructed Wetland System 1 (*Schoenoplectus/Scirpus/Juncus*) and Wetland System 2 (*Typha/Sparganium*).**

Cell	Depth	Mean <sup>†</sup>	SD
<b>System 1</b>			
1	0-5	7.08	0.13
1	5-10	7.21	0.22
1	10-15	7.13	0.17
1	15-20	7.05	0.08
2	0-5	7.17	0.16
2	5-10	7.25	0.27
2	10-15	7.13	0.15
2	15-20	6.95	0.11
<b>System 2</b>			
3	0-5	7.03	0.03
3	5-10	7.05	0.09
3	10-15	6.79	0.03
3	15-20	6.48	0.20
4	0-5	7.09	0.12
4	5-10	6.94	0.18
4	10-15	6.43	0.21
4	15-20	6.25	0.02

<sup>†</sup> Mean of 4 yr.



**Fig. 4. Relationship of mean  $\text{NH}_4^+\text{-N}$  inflow concentration vs. mean  $\text{NH}_4^+\text{-N}$  pore water concentration (pooled by depth) of each wetland cell for Wetland System 1 (WS1, *Schoenoplectus/Scirpus/Juncus*) and Wetland System 2 (WS2, *Typha/Sparganium*).**

for Wetland System 1 were consistently higher than for Wetland System 2 (Hunt et al., 2002). In the present study, Wetland System 1 (bulrush/rush) had lower mean  $\text{NH}_4^+\text{-N}$  concentrations in pore water than Wetland System 2 (cattails/bur-reed) when  $\text{NH}_4^+\text{-N}$  inflow levels were  $>70 \text{ mg L}^{-1}$  (Fig. 4). Therefore, bulrush/rush wetlands had 34 to 60% higher  $\text{NH}_4^+\text{-N}$  fluxes from the soil into the wastewater than the cattails/bur-reed wetlands, probably because their plant-soil environment provided conditions for more efficient microbial removal of N.

#### Wastewater N loading Rates and $\text{NH}_4^+\text{-N}$ Concentration

A major factor controlling the  $\text{NH}_4^+\text{-N}$  accumulation in pore water was  $\text{NH}_4^+\text{-N}$  concentration in wastewater. Consequently, mean  $\text{NH}_4^+\text{-N}$  concentration in pore water was highly correlated with mean  $\text{NH}_4^+\text{-N}$  concentration in inflow of each cell in both systems (Fig. 4). These results show that the soil pore water  $\text{NH}_4^+\text{-N}$  levels increased with higher  $\text{NH}_4^+\text{-N}$  concentration in applied wastewater. As the total N-loading rates increased annually in both wetland systems (Table 2), soil appears to be storing more and more of the  $\text{NH}_4^+\text{-N}$  in the soil column. This was most dramatic in Wetland System 2, Cells 3 and 4 (Fig. 3). As the soil pore water profile became saturated with  $\text{NH}_4^+\text{-N}$ , the N-removal capacity of the wetlands decreased. Mass removal efficiencies dropped below 75% when loading rates were  $>2.5 \text{ gm}^{-2} \text{ d}^{-1}$  (Stone et al., 2002). These high loading rates coincide with inflow  $\text{NH}_4^+\text{-N}$  concentrations  $>200 \text{ mg L}^{-1}$  (Fig. 4). Most likely, accumulation of high levels of  $\text{NH}_4^+\text{-N}$  in the soil column will negatively affect both sustainability and long-term ability of the wetland systems to treat wastewater with high levels of N. As indicated by Humenik et al. (1999), additional pretreatment

such as wastewater aeration and nitrification will be required to reduce the negative effects of high  $\text{NH}_4^+$ -N concentrations.

## CONCLUSIONS

This study evaluated N storage and distribution in soils of two constructed wetland systems that treated swine lagoon wastewater for 5 yr at increasing loading rates of N. The 5-yr net total N accumulation was not significantly different ( $P < 0.10$ ) between Wetland System 1 ( $106 \text{ g m}^{-2}$ ), with a bulrush/rush plant community, and Wetland System 2 ( $100 \text{ g m}^{-2}$ ), with a cattail/bur-reed community. Both wetland systems did not show significant difference in soil N storage pattern by depth. On average for both systems, the upper 5-cm layer stored 34% of the total soil N. Although total soil N accumulation significantly increased with time in both systems, constructed wetland soils stored small amounts of N with respect to the total applied N (<6% of total N applied during the 5 yr).

Profiles of  $\text{NH}_4^+$ -N distribution in soil pore water showed that at N loading rates of  $0.6 \text{ g m}^{-2} \text{ d}^{-1}$ ,  $\text{NH}_4^+$ -N was transported from the surface water into soil pore water following a negative diffusive flux across the soil-water interface. This downward movement suggested that  $\text{NH}_4^+$ -N moved to satisfy the lower concentration gradient in lower parts of the soil profile. At higher loading rates, 1.2 to  $2.7 \text{ g m}^{-2} \text{ d}^{-1}$ , the profile showed that  $\text{NH}_4^+$ -N was transported upwards, from the soil pore water into the surface water. This upward flux suggested that microbial nitrification and denitrification were probably the major removal processes at the water-soil interface. Bulrush/rush wetlands had higher  $\text{NH}_4^+$ -N fluxes from soil into wastewater than cattails/bur-reed wetlands, probably because their plant-soil environment provided conditions for more efficient microbial removal of N.

A major factor controlling the  $\text{NH}_4^+$ -N accumulation in pore water was  $\text{NH}_4^+$ -N concentration in lagoon wastewater. In both systems, mean  $\text{NH}_4^+$ -N concentration in pore water was highly correlated with mean  $\text{NH}_4^+$ -N concentration in the inflow of each cell. As the total N loading rates increased annually in both wetland systems, soil pore water had higher levels of  $\text{NH}_4^+$ -N but N removal efficiency of the wetlands sharply decreased. Accumulation of high levels of  $\text{NH}_4^+$ -N ( $>200 \text{ mg L}^{-1}$ ) in the soil pore water column may negatively affect both plant growth and long-term ability of wetland systems to treat wastewater with high N levels. Additional pretreatment such as wastewater aeration and nitrification may be required to reduce the negative effects of high  $\text{NH}_4^+$ -N concentrations.

## ACKNOWLEDGMENTS

This research was partially funded by N.C. Herrings Marsh Run Water Quality Demonstration Project, USDA Project No. 90-EWQD-1-9504, N.C. Goshen Swamp Hydrologic Unit Area Project, USDA Project No. 90-EHUA-1-0013 and USEPA Project No. CR823808-01-0 "Evaluation of Alternative Constructed Wetland Systems for Swine Wastewater

Treatment". The authors thank T.A. Matheny, J.M. Rice, and V.C. Rogers for technical assistance.

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