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NITROGEN TRANSFORMATIONS IN A SIMULATED OVERLAND FLOW WASTEWATE--ETC(U)
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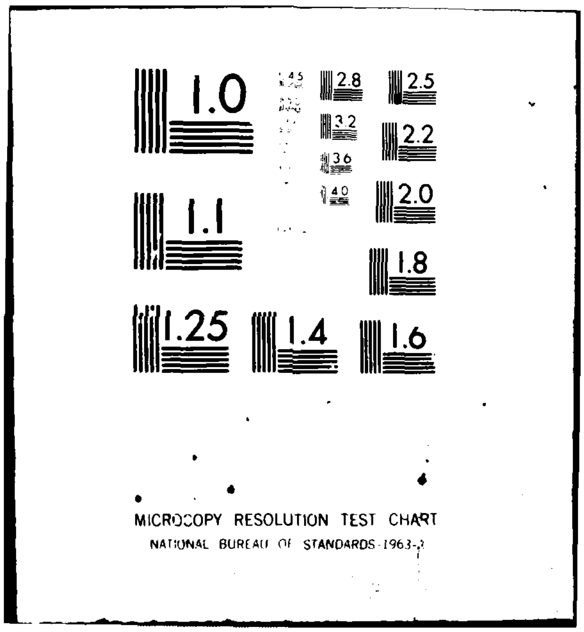
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NITROGEN TRANSFORMATIONS IN A SIMULATED OVERLAND FLOW WASTEWATER TREATMENT SYSTEM.

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Treating wastewater in properly designed and operated overland flow systems results in significant amounts of N being removed through nitrification-denitrification reactions. Application of wastewater containing NH_4^+-N in a simulated overland flow model led to the disappearance of ammonium and the formation of nitrate in oxidized surface soil. The N balance in the simulated overland flow system was estimated by using labeled ^{15}N. The amount of N removed in the system depends upon denitrification rates. The results of this

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study indicated that N adsorption on the soil complex and uptake of applied ammonium by vegetation accounted for the N removed in the overland flow systems. The adsorbed ammonium on the aerated surface soil mass was nitrified and converted to oxidized forms of N. The nitrate thus formed diffused downward to the reduced zone during subsequent wastewater applications. Some of this nitrate then denitrified and converted to gaseous forms of N or was assimilated and reduced by plant life. Results of the overland flow studies indicated that approximately 55-68% of wastewater NH_4^+-N added to the simulated overland flow system was unaccounted for in controlled laboratory environments. cro ~~This NH_4^+-N~~ was presumably returned to the atmosphere.

NH4(+)-N

PREFACE

This report was prepared under contract to the U.S. Army Cold Regions Research and Engineering Laboratory by R.L. Chen, Water Chemist, Waterways Experiment Station, Corps of Engineers, Vicksburg, Mississippi and William H. Patrick, Jr., Center for Wetland Resourses, Louisiana State University, Baton Rouge, Louisiana. The report was funded under Civil Works Program, Wastewater Management, Research Sub-program, Land Treatment, Work Unit, CWIS 31634.

Dr. Y. Nakano and Dr. Alex Iskandar of USACRREL reviewed the report. Dr. Nakano also acted as technical monitor.

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INTRODUCTION

Overland flow treatment of wastewater has gained favor as an effective economical method of removing nutrients and pollutants from wastewater as is required by the Federal Water Pollution Control Act. Although land treatment of municipal and industrial wastewater has been successful in several locations in the world, few researchers have studied the effects of the treatment on transformation rates and N removal efficiency.

Overland flow treatment of wastewater could improve wastewater quality to meet minimum water quality standards proposed by the EPA for effluent before it is discharged into surface waters. In a conventional overland flow treatment system, wastewater is applied to grassed slopes and allowed to flow through the vegetative litter as a film of water over the surface of impermeable soil. Research on overland flow wastewater treatment has indicated that the effluent flowing over the land surface would lose a considerable amount of nutrients.

The overland flow treatment of wastewater produces conditions similar to those occurring in rice soils which are alternately flooded and dried. The process has a profound effect on normal soil processes. Below the oxidized surface soil, the absence of oxygen results in a reduced zone characterized by low redox potential. Thus, both oxidation and reduction occur in an overland flow wastewater treatment system. Oxidized conditions favor the conversion of applied NH_4^+-N to NO_3^--N (nitrification). The NO_3^--N may diffuse down to the lower anaerobic zone or be taken up by the growing vegetation. The absence of O_2 in the anaerobic zone favors denitrification, nitrate reduction, and N mineralization. If both oxidized and reduced conditions are found at the soil/water interface, the tertiary treatment of wastewater by land application may increase N removal, improving water quality.

Research has been conducted in recent years to characterize aerobic and anaerobic zones that affect the rates of nitrogen transformation reactions. The thickness of the aerobic layer is reported to vary from a few millimeters to approximately 1 cm (in quiescent submerged soil or lake sediment). The degree of biological activity depends on the organic material contained in the system (Mortimer 1941, 1942, Lee 1970, Patrick and DeLaune 1972, Engler and Patrick 1974). The oxidation-reduction conditions at the water/soil interface have a major impact on the N balance in submerged environments (Mortimer 1971, Keeney 1973). Unless the overlying water is completely anoxic, simultaneous nitrification-denitrification should occur at the water/soil interface (Lee 1970, Keeney 1973, Chen et al. 1979).

Goering and Dugdale (1966), Chen et al. (1972), Brezonik (1973) and Payne (1976) have discussed the role of denitrification in converting NO_3^- to gaseous forms of N under various redox and pH conditions. The rate of denitrification is regulated by the Eh and pH conditions in the system.

The nitrogen budget in a waterlogged system was estimated recently with a stable isotope tracing technique. Catchpool (1975) reported that gaseous losses of applied ^{15}N on a Rhodes-grass pasture-field microplot were as high as 27% when the soil was waterlogged. Approximately 50% of the applied NH_4^+-^{15}N was lost through simultaneous nitrification-denitrification reactions in a simulated overland wastewater treatment system (bare soil system) (Chen and Patrick 1978). Nitrogen disappearance is significant in flooded soils and possibly in marshes and wetlands (Ponnamperuma 1972, Isirimah and Keeney 1973, Spangle et al. 1976). Nitrogen removal in an overland flow system depends on soil type and N application rates, and shows pronounced seasonal variations (Thomas et al. 1976). The amount of N taken up by plants also has been estimated in laboratory and field studies (Chen and Patrick 1977, Peters and Lee 1978, Iskandar et al. 1976). Carlson et al. (1974) reported that 31% of the N applied in a secondarily treated municipal wastewater was removed by grass sod in an overland flow test model with a 2% slope. In a simulated wastewater application system, Chen and Patrick (1978) found that approximately 12 to 20% of the labeled NH_4^+-^{15}N applied was taken up by plants in a Mhoon soil-ryegrass system. Khalid et al. (1978) found that a similar quantity (16.5%) of ^{15}N applied was incorporated into grass in an Olivier-soil ryegrass system.

Our research employed scale models of plant-soil systems in which labeled ^{15}N (as NH_4^+-N) was used to trace applied N during overland flow. The N removal efficiency and the amount of applied N incorporated in the plant-soil system were of special interest.

EXPERIMENTAL PROCEDURES

Description of soil used

A low permeability Mhoon silt loam soil was collected from the USDA Ben Hur Research Farm at Baton Rouge, Louisiana. The properties of the soil are listed in Table 1.

Table 1. Properties and particle size distribution of Mhoon soil used for the overland flow experiment.

Sand	Silt	Clay	CEC	pH	NH_4^+	NO_3^-	Org N
----------	%---------		meq/100 g soil		--- µg	N/g soil	---
43.0	42.9	13.1	11.0	5.8	4.4	5.2	857.4

The soil samples were air-dried and passed through a screen with 6.3-mm openings. The samples were thoroughly mixed and stored in sealed plastic-lined containers prior to use. Moisture content of the soil was determined when the soils were packed in the overland flow test models. Twenty-two milliequivalents of $CaCO_3$ was added to the 100 g of soils and mixed thoroughly to raise the soil pH to 7.1, providing favorable conditions for N transformations (Broadbent and Clark 1965).

Description of the overland flow test model

The container used for the overland flow experiment was constructed of 1.90-cm-thick plywood with inside dimensions of 118 cm L x 16.2 cm W x 15 cm D. The interior of the test container was strengthened with fiber glass cloth and painted with several coats of polyurethane to prevent water seepage. A Plexiglas water collection trap at the top of the lower end of the model collected runoff. A drainage port at the bottom of the same end of the container collected subflow. A longitudinal section of the test model is shown in Figure 1.

Approximately 23 kg (oven-dried weight) of limed Mhoon soil was uniformly packed into the test model. The soils were compressed by hand with hardwood blocks to attain a density similar to natural conditions.

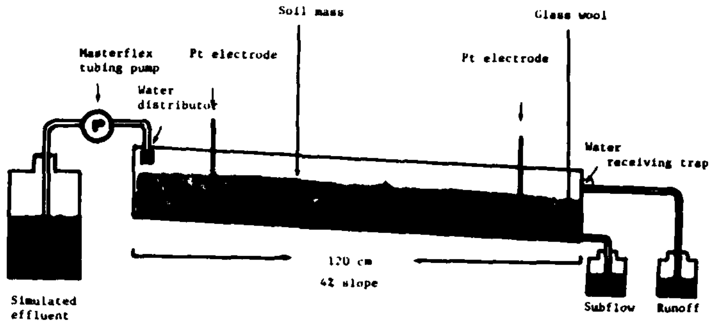

Figure 1. Schematic diagram of the overland flow wastewater treatment model.

Soil depth in the test model was 10 cm. Glass wool was placed across the width of the lower end to provide drainage to the subflow outlet.

Perennial ryegrass (Lolium perenne L.) was seeded at the rate of 100 g/m^2 and watered occasionally with tap water until it germinated. The model was maintained for 4 weeks by applying simulated wastewater to establish the sod. The models were placed on a 4% slope and put in a constant temperature room (20° to 22°C) fitted with Agro-Lite and fluorescent lights for normal plant growth. Simulated waste effluent was applied to models at the upper end using a multi-channel tubing pump (Cole-Palmer, model 7567). The simulated wastewater contained NH_4^+-N solution, dextrose, and a plant nutrient solution. It was applied at a rate of 1.57 cm^3/cm^2-day (equivalent to 3 liters/day-model). This volume of wastewater was applied over an 8-hour (Treatments A, B, C, and E) or a 16-hour (Treatment D) period. Applications were made on five consecutive days per week for A, B, C, and E treatments and three alternate days per week for Treatment D during the entire study period. The treatments are summarized in Table 2. Runoff and subflow samples were collected on each day of application and analyzed for inorganic N within 24 hours. Organic N (TKN) was measured periodically.

Table 2. Methods of wastewater application.

Treatment	NH_4^+-N conc. in wastewater (μg N/ml)	Application period (hr)	Application schedule (days/wk)
A	25	8	5 consecutive
B	50	8	5 consecutive
C	75	8	5 consecutive
D	75	16	5 consecutive
E	50	8	3 alternate

Various concentrations of ammonium-N (as $(NH_4)_2SO_4$) were added to the simulated wastewater to give total N contents of approximately 25, 50, and 75 μg N/ml. Average flow rates were either 3.13 or 6.25 ml/min, depending upon the treatment. At the start of wastewater applications, one application contained the labeled NH_4^+-^{15}N solution as a tracer.

Overland flow and subflow effluent samples were taken on each application day and analyzed for NH_4^+, NO_3^-, and NO_2^- concentrations. The effectiveness of wastewater N removal in an overland flow system was assessed by determining the rate of N disappearance from the effluents. Redox potentials in the soil profile were measured every day at three different soil depths at two different locations downslope to monitor the redox conditions of the soil-plant system during the investigation.

The ryegrass was trimmed to a height of 5 cm before simulated wastewater containing labeled N was applied to the test models. Second and third grass harvests were made 8 and 37 days after the application of the effluent containing labeled N. The plant samples were air-dried at 55°C and their TKN determined. The experiment ended after 40 days. At the end of the experiment, the grass was separated from the soil and the soil mass was removed from the containers and divided into 30-cm cross sections. Each 30-cm section was divided into three equal layers

(3.3 cm) to determine the concentrations and forms of N in the soil. The distribution of ^{15}N in various forms in the soil, effluent, and plant samples were measured on a DuPont mass spectrometer (model 21-621) using the NaOBr method.

Chemical analyses

Water samples were placed in tightly sealed glass bottles containing a few drops of concentrated H_2SO_4 as a preservative and were stored at 4°C in the dark prior to analyses. Chemical procedures were as follows: NH_4^+ and NO_3^--N were determined by steam distillation with MgO and Devarda alloy (Bremner and Keeney 1965) followed by either Nessler's reagent method (EPA 1974) or acid titration. Nitrite-N concentration was estimated by a modified Griess Ilosvay method (Bremner 1965b), total N by the semi-micro Kjeldahl procedure (Bremner 1965a), and exchangeable NH_4^+-N by 2 $\underline{M}$ KCl extraction and steam distillation (Bremner and Keeney 1966). Effluent and soil (soil-water ratio = 1:1) pH were determined with a glass electrode and the specific conductivity (corrected to 25°C) was measured with a mho-meter (YSI, model 33) equipped with a conductivity probe.

Redox potential measurements

Redox electrodes were constructed by connecting a 20-gauge Pt wire to a 16-gauge Cu wire in a glass tube filled with Hg. A 3% KCl salt bridge was used to make the connection between the soil system and a calomel reference electrode. A Beckman zeromatic meter (model SS-3) was used for all Eh and pH measurements. Vertical distribution of Eh was measured with the technique developed by Patrick and DeLaune (1972).

The Pt electrode was adjusted and driven downward through undisturbed soil in the upslope end of the test model at a constant rate of 2 mm/hr. An Orion pH meter (model 407) coupled with an OmniScribe recorder was used for Eh profile measurements.

Nitrogen-15 analyses

Nitrogen-15 analyses were conducted on distillates obtained from total N and inorganic N analyses. The distillates were collected in a 100-ml beaker and a few drops of 1 M H_2SO_4 were added. The samples were then condensed to about 2 ml in an oven at 70°C. Nitrogen-15 isotope ratio analyses were performed on a DuPont mass spectrometer (Model 21-621) using the NaOBr method (Bremner 1965c). Sample cross-contamination was eliminated by using the 90% alcohol double distillation procedure (Bremner and Edwards 1965).

RESULTS AND DISCUSSION

Water budgets

The runoff and subflow effluents were collected and the volumes of effluent were measured each day of wastewater application to estimate the recovery rate of wastewater. Runoff and subflow effluents were recovered with approximately a 1 to 2 ratio by volume at the end of the experiment.

In 8-hour application treatments, only 46% of the applied effluent was recovered after the 2-day break in water application. Low recovery of applied effluent was anticipated in this relatively dry soil because of its water retention capacity. Recovery of effluent then increased and remained constant at an average rate of 75% for the rest of the week.

The overall recovery rate of applied wastewater from the test model was approximately 69% (Table 3). This suggests that 31% of the applied water, equivalent to a 0.50-cm depth, either evaporated from the soil surface or evapotranspired from the grass.

In the longer application period (16 hrs/day) study (Table 3), 46% of the wastewater applied to the overland flow model was recovered in the first day of the weekly application cycle. Water recovery increased to approximately 70% and remained steady in the rest of the applications. The average water recovery rate was 65% for the 16-hour application schedule.

The rate of nitrogen removal in an overland flow system is primarily controlled by nitrification-denitrification reactions. A treatment of three applications on alternate days per week (Treatment E) was designed to facilitate nitrification by extending the drying period in the overland flow system. Lower water recovery from this system was expected due to greater evaporation and plant uptake. At the end of the experiment an average of only 54% of the applied wastewater was recovered from the system.

Redox potential measurements

The redox potentials were monitored at two locations once every application day at depths of 1, 6, and ? cm (Fig. 1). The redox potentials of the soil-plant system 1 cm from the surface ranged from +300 to +500 mV throughout the experiment (Table 4). The soil Eh of +400 mV measured at the surface by the upslope electrode suggested that a portion of NH_4^+-N added to the overland flow test model may have undergone nitrification (Ponnamperuma 1972). The Eh of the soil at the downslope electrode ranged between +200 and +300 mV for the first 30 days of the experiment, but fluctuated between +100 and +450 mV thereafter. Since Eh readings greater that about +350 mV indicate conditions that favor nitrification, it is likely that the surface layer of the soil-plant system provided favorable conditions for oxidation of NH_4^+-N retained by the soil mass. The redox potential values of the middle layer (6 cm from the surface) remained under +200 mV. Most readings were less than 0 mV. The Eh of the bottom soil layer (9 cm depth) remained consistently low, ranging from 0 to -300 mV. Because of the reduced conditions in the bottom soil, it can be assumed that nitrate diffusing down from the surface layer was denitrified or assimilatorily reduced. The amount of nitrate that might have leached out from the overland flow test model depended on the rate of denitrification and assimilatory nitrate reduction in the system.

In the 16-hour application model, the surface layer upslope remained in an oxidizing state throughout the entire study. However, the Eh of the soil indicated that reducing conditions existed at the downslope region. In the bottom soil layer (depth > 6 cm) the Eh level was lower than -100 mV, indicating very strong reducing conditions (Table 5). In the three alternate days per week treatment, the redox potentials of the surface layer ranged from +350 mV to +550 mV and remained strongly oxidizing throughout the investigation (Table 6). The bottom soil layer remained in a reducing state during most of the experiment, but lost some reducing potential toward the end of the experiment.

The vertical distribution of redox potentials of the soil-plant system was also measured during the overland flow experiment. Redox measurements when wastewater was applied for 8 hours showed that the Eh of the surface soil ranged from +300 to +400 mV or higher, while the Eh of the bottom soil remained below 0 mV (Fig. 2). The Eh of the bottom soil increased by approximately 100 mV during wastewater application, indicating that NO_3^--N was diffusing downward from the surface layer. The vertical distribution of redox potentials in the soil-plant system with a 16-hour application period (Treatment D) also showed that only a thin layer of surface soil retained oxidizing potential. The Eh of the soil deeper than 1 cm declined drastically from +300 mV to approximately 0 mV within 1 cm, and then declined gradually through the soil profile. In most of the treatments, the bottom soil Eh remained at -150 to -200 mV throughout the entire investigation.

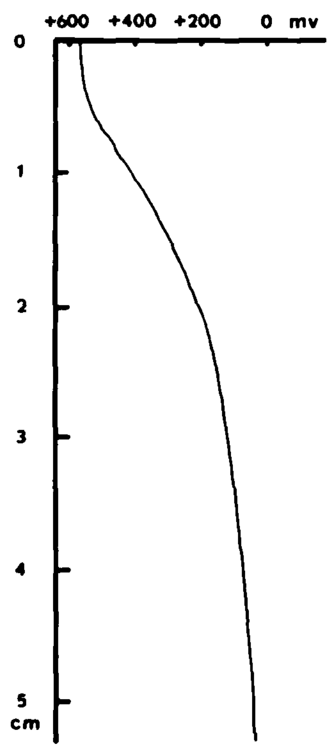

Figure 2. Vertical distribution of Eh in an overland flow wastewater treatment model.

Redox potential levels in the overland flow model were affected substantially by the addition of 100 mg C/liter as an energy source. Greater reducing conditions developed and a higher rate of denitrification existed in the system after the energy source was added.

The Eh data presented here are not corrected to a standard pH. The pH of the system during the investigation averaged 7.1 in the overall system and varied by ± 0.5 at most. There was no indication of redox electrode poisoning (Bailey and Beauchamp 1971) in most readings.

Efficiency of nitrogen removal

Data summarized in Table 7 indicate that NH_4^+ and NO_3^--N were released from the soils. Nitrogen removal from simulated wastewater was about 88% in water containing 25 μg N/ml in NH_4^+ form (Treatment A). Lower removal rates ranging from 75 to 85% were found in the other treatments. The addition of 100 mg C/liter as dextrose to the simulated wastewater increased N removal by 10%. Approximately 98% of the applied N was removed from the effluent in the 25-μg N/ml treatment with C addition. The percentage of N removed in the soil-plant system declined as NH_4^+ concentrations in the simulated wastewater increased. Approximately 10 and 19% of the added NH_4^+-N was recovered in the effluent in the 50- and 75-μg N/ml of NH_4^+-N treatments (Treatment B and C) respectively, with an 8-hour application period. Analysis of inorganic N in the effluent showed that approximately 10 to 15% of the added NH_4^+-N was oxidized and converted to the NO_3^- form, while 1.1 to 16.5% of the added NH_4^+-N was recovered from the effluent in Treatment E, in which applications were made on alternate days. In treatments C and D

(75 μg N/ml of NH_4^+-N) inorganic N concentrations in the effluent decreased as the application time increased from 8 to 16 hours. Only 13% of the added NH_4^+-N was recovered in inorganic forms in Treatment D, compared with 19% in Treatment C. Periodic determinations of TKN contained in the effluents indicated that the concentration of organic N was usually low and remained under 4 μg N/ml. Concentrations of organic N in the effluents decreased with time. On the average, 2% of the added NH_4^+-N remained as NO_3^--N and 10% of the added N remained as NH_4^+-N in the effluent from an 8-hour application.

Nitrification apparently was not affected in an overland flow system when dextrose was added. Nitrate distribution in the effluent showed that C added to applied wastewater resulted in higher nitrate reduction rates after a three day lag period. The N removal rate in the effluent increased with increased water residence time. Nitrogen removal was increased from 80% to 87% when the wastewater application time was increased from 8 to 16 hours in the 75-μg N/ml treatment. The longer application time favored a higher rate of N removal in the overland flow system.

Distribution of added NH_4^+ - ^{15}N in the effluents

The distributions of added NH_4^+-^{15}N in the effluents are shown in Table 8. Quantitative analysis of ^{15}N in the effluents showed that 10.4 to 29.9% of the applied NH_4^+-^{15}N was discharged from the system during the experiment by the various treatments. The labeled N recovered indicated that the applied wastewater contributed most of the NH_4^+-N in the effluent in most of the treatments. The recovery of labeled N in the effluent decreased drastically over time as subsequent applications of unlabeled N diluted the labeled N. Approximately 90 to 95% of the NH_4^+-^{15}N discharged in the effluent was recovered within 5 days after NH_4^+-^{15}N was applied. Similarly, 95% of the total NO_3^--^{15}N discharged was recovered within five days after wastewater containing NH_4^+-^{15}N was applied.

In treatment C (75 μg N/ml NH_4^+-N, 8-hour application), approximately 14% of the added NH_4^+-^{15}N was recovered within a day after the ^{15}N-labeled wastewater was applied, while only 2% of added NH_4^+-^{15}N was recovered from treatment D (16-hour application period) within a day. Ammonium was rapidly converted to oxidized forms of N in the early part of the experiment. With C added, only 2% of the added NH_4^+-^{15}N was released in NO_3^- form from the soil model within eight hours of application. Application of wastewater containing 100 μg C/ml as dextrose to an overland flow system resulted in a noticeable increase in the rate of NO_3^- disappearance. Lance and Whisler (1976) and Sikora and Keeney (1976) have reported the enhancement of denitrification by carbon addition in a laboratory incubation of soil.

Distribution of added NH_4^+-^{15}N in the soil model

The distribution of ^{15}N added to the soil was found by determining the concentrations of various N forms at various distances down slope and at various depths at the conclusion of the study. The soil mass was removed from the test container and divided into 30-cm sections. The concentration gradients of ^{15}N in the soil beds are presented in Tables 9-13. Nitrogen-15 analysis of the soil N showed that a major portion of the retained ^{15}N was incorporated into the soil's organic fraction.

With the exception of treatment D, less than 1% of ^{15}N was recovered from the soil in inorganic forms. Ammonium-N added to the soil was adsorbed by the soil complex immediately. The adsorbed NH_4^+-N on the soil complex was initially converted to NO_3^--N in the surface-oxidation zone, and then taken up by growing vegetation or immobilized by soil microbes and incorporated into organic forms. Because of its mobility in the soil system the nitrate thus formed diffused to the reduced zone of the soil mass when wastewater application resumed. Some of the nitrate was denitrified to gaseous forms of N in the reduced zone, evolved through the soil mass, and was lost to the atmosphere. The rate of nitrate disappearance was increased by the addition of C. The concentration of NO_2^--N in the soil column was normally undetectable. The distribution of labeled N in the soil profile showed that the added NH_4^+-^{15}N accumulated in the surface soil within 30 cm of the upslope end. Further than 30 cm from the application port, very little added NH_4^+-^{15}N accumulated. The distribution of NO_3^--^{15}N in the soil column was similar to that of NH_4^+-^{15}N except in treatment D. Retention of labeled N in the soil column was similar to that of NH_4^+-^{15}N except in treatment D. Retention of labeled N in the soil columns increased significantly when the application time was increased from 8 hours to 16 hours. The high concentration of NO_3^--^{15}N at the bottom of the soil profile was possibly due to nitrification that occurred after the wastewater applications terminated but prior to analysis.

The amount of added NH_4^+-^{15}N incorporated into the organic fraction decreased with downslope distance and was several times greater than that of inorganic forms. Approximately 4.2 to 9.7% of the added NH_4^+-N was adsorbed by the soil complex and transformed into organic forms at the end of the experiment.

Removal of added NH_4^+-^{15}N through plant uptake.

To estimate the rate of ^{15}N uptake by vegetation in the different treatments, the grass was harvested and samples were analyzed. The ryegrass plants were cut down to a height of 5 cm before the labeled NH_4^+-^{15}N was introduced to the overland flow test models. The concentrations of ^{15}N in the ryegrass are presented in Table 14. Nitrogen-15 mass balance calculations indicated that uptake of ^{15}N by ryegrass accounted for 11.4 to 21.8% of the added NH_4^+-^{15}N. No symptoms of N deficiency were shown in the ryegrass in the overland flow test model. The distribution of ^{15}N in ryegrass growing at various distances downslope indicates that the rate at which N is taken up is nonlinear with distance (Table 15).

Nitrogen balance in Mhoon soil ryegrass overland flow test model

Nitrogen balance of the simulated overland flow system under various treatments was estimated after wastewater had been applied for 32 days. The concentrations and distributions of organic and inorganic N in effluent, soil complex, and plant samples are shown in Tables 16-20.

The data indicate that from 10 to 30% of the added NH_4^+-^{15}N was recovered in the effluent. In contrast to our previous experiment of bare soil systems (Chen and Patrick 1977), a greater portion of the recovered inorganic N was in the $(NO_3^- + NO_2^-)$-N form, accounting for 8 to 11% of the recovered NH_4^+-^{15}N. The amount of the added ^{15}N recovered in the effluent, present as NH_4^+-^{15}N in the effluent decreased from 21 to 13% when wastewater application time was increased from 8 to 16 hours. This confirmed the importance of residence times in the overland flow treatment system. Extension of the resting period between wastewater applications did not enhance N removal rates. More added NH_4^+-^{15}N was discharged and recovered in the effluent of treatment E (three alternate days of wastewater application) than in the other treatments (five consecutive days of application). Nitrogen removal by ryegrass in the overland flow test model was about 11.4 to 21.8% of the applied NH_4^+-^{15}N. Approximately 18 to 22% of the added NH_4^+-^{15}N was taken up by plants in the low and medium N treatments (25 to 50 μg N/ml). Only 11 to 13% of the added NH_4^+-^{15}N was taken up from the high N treatment (75 μg N/ml). In contrast to the bare soil system, which retained 35% of the added NH_4^+-^{15}N in the soil complex (Chen and Patrick 1977), only about 4.5 to 9.8% of the applied NH_4^+-^{15}N was recovered in the soil complex at the end of the 32-day study. Nitrogen budget calculations for the overland flow test model showed that approximately 55 to 68% of the added NH_4^+-^{15}N was unaccounted for under various N treatments and was therefore presumably lost to the atmosphere. The loss of applied ^{15}N was attributed mainly to denitrification and possibly ammonium volatilization. Since denitrification and NH_3 volatilization were not estimated in the overland flow system, more research on these topics is urgently needed in order to provide precise quantitative data on N transformations in this system.

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Table 3. Distribution of simulated wastewater applied to the mhoon soil-ryegrass system with 4% slope at 22°C.

	Average water recovery (%)								
	Treatment A, B, C			Treatment D			Treatment F		
	Overland runoff	Sub-flow	Combined outflow	Overland runoff	Sub-flow	Combined outflow	Overland runoff	Sub-flow	Combined outflow
1st day (Mon)	13.4	32.3	45.9	18.8	26.9	45.7	19.7	28.0	47.7
2nd day (Tue)	29.4	45.6	75.0	24.1	42.6	66.7	—	—	—
3rd day (Wed)	24.7	50.2	74.9	16.7	53.8	70.5	19.2	37.7	56.9
4th day (Thu)	23.3	52.3	75.5	19.4	53.0	72.4	—	—	—
5th day (Fri)	25.0	50.7	75.7	30.6	39.3	69.9	21.4	35.9	57.3
Average	23.2	46.2	69.4	21.9	43.1	65.0	20.1	33.9	54.0

Table 4. Effect of soil depth and model distance on redox potential (Eh, mv) with time in mhoon soil-ryegrass model in an overland flow treatment system with an 8-hour wastewater application period at a 4% slope.

Date		Distance downslope, cm			
	20 cm			100 cm	
	1 cm	6 cm	9 cm	1 cm	6 cm
March 21	252	29	-185	70	-132
22	258	8	-215	56	-147
23	313	10	-220	95	-144
24	277	-8	-235	114	-153
25	284	-33	-225	115	-155
27	355	-36	-225	140	-117
28	343	-25	-225	142	-139
29	305	-16	-210	83	-161
30	339	-39	-240	53	-190
31	406	-37	-225	63	-174
April 2	419	55	-255	116	-158
3	383	13	-225	104	-155
4	409	-12	-235	72	-180
5	409	-44	-250	15	-195
6	327	-68	-250	-5	-215
7	437	-53	-235	80	-192
9	434	-78	-230	107	-170
10	449	-8	-230	168	-66
11	371	-96	-245	26	-194
12	396	-103	-235	39	-191
13	293	106	-245	48	-194
14	421	-117	-245	32	-208
17	444	13	-130	187	-90
18	392	-115	-220	85	-179
19	326	114	-235	35	-200
20	346	115	-230	35	-200
21	317	-101	-200	54	-191
22	317	-111	-220	91	-177
24	452	91	-185	279	-138
25	360	1	-85	138	-19
26	432	15	-155	153	-31
27	367	32	-135	112	-43
28	399	41	-155	92	-53
29	394	-24	-135	47	-97
30	454	105	-185	167	7
May 1	420	111	-175	303	104
2	367	10	-190	138	2
3	293	5	-155	44	-83
4	337	-20	-155	44	-131
5	448	-35	-155	114	-112
6	440	-25	-135	83	-152
7	501	43	-260	165	42
8	465	48	-270	250	172
9	429	-10	-175	147	-28
10	326	0	-185	107	-43
11	334	-15	-170	122	-63
12	478	15	-245	138	-64
13	499	-10	-260	134	-13
15	478	78	-210	377	262
16	299	105	-275	242	124
17	428	110	-295	300	162
18	471	100	-245	247	138
19	427	100	-230	291	157
20	481	98	-260	300	143
21	477	103	-150	393	287
22	300	170	-10	396	289
23	336	163	-85	310	246
24	371	155	-62	395	266
25	490	173	-75	442	299
26	492	158	-20	391	304
29	480	315	115	474	441

Table 5. Effect of soil depth and model distance on redox potential (*Eh*, mv) with time in mhoon soil-ryegrass model in an overland flow treatment system with a 16-hour wastewater application period at a 4% slope.

Date	Distance downslope, cm: 20 cm			100 cm	
	1 cm	6 cm	9 cm	1 cm	6 cm
March 21	315	-150	-245	315	-93
22	340	-173	-260	293	-118
23	355	-168	-255	320	-113
24	368	-188	-265	315	-120
25	370	-195	-265	310	-88
27	470	-198	-260	325	-95
28	385	-205	-265	345	-113
29	410	-210	-265	360	-130
30	375	-200	-295	293	-135
31	415	-225	-285	355	-140
April 2	460	-213	-265	293	-193
3	410	-198	-275	310	-155
4	445	-235	-265	330	-168
5	405	-195	-285	323	-178
6	375	-210	-280	333	-190
7	468	-220	-265	323	-173
9	488	-213	-260	353	-160
10	498	-125	-255	333	-170
11	438	-200	-275	330	-125
12	445	-158	-265	330	-170
13	450	-185	-265	343	-163
14	405	-238	-265	330	-170
17	423	-88	-175	355	-180
18	410	-155	-260	375	-165
19	420	-125	-255	370	-195
20	395	-200	-255	360	-170
21	413	-183	-255	360	-173
22	408	-85	-245	370	-178
24	410	-5	-115	415	-175
25	335	-8	-35	370	-168
27	470	-123	-175	370	-145
28	350	-210	-220	368	-148
29	350	-220	-235	300	-160
30	340	-60	-240	370	-155
May 1	365	-50	-235	363	-185
2	335	-105	-235	345	-160
3	235	-140	-185	358	-160
4	265	-70	-225	375	-158
5	305	-90	-225	320	-133
6	240	-123	-225	370	-150
7	325	-15	-220	365	-155
8	320	-15	-230	355	-180
9	260	-68	-235	375	-140
10	275	-195	-230	345	-148
11	260	-120	-230	355	-148
12	315	-135	-235	325	-175
13	278	-150	-225	335	-148
15	370	5	-125	355	-170
16	275	-45	-185	320	-160
17	348	-30	-200	345	-160
18	295	-70	-215	365	-155
19	370	-43	-220	345	-163
20	235	-73	-225	340	-148
21	420	-23	-10	360	-168
22	338	0	-20	355	-160
23	283	25	-100	320	-155
24	289	20	-105	275	-138
25	270	-25	-105	345	-158
26	255	-50	-115	355	-148
29	418	283	-125	360	-165

Table 6. Effect of soil depth and model distance on redox potential (Eh, mv) with time in mhoon soil-ryegrass model in an overland flow wastewater treatment system with an 8-hour wastewater application period 3 alternative days per week at a 4% slope.

Date	Distance downslope, cm: 20 cm, 1 cm	20 cm, 6 cm	100 cm, 1 cm	100 cm, 6 cm
March 21	243	-35	340	-45
22	270	-78	330	-68
23	280	-80	350	-58
24	283	-115	353	-75
25	300	-135	373	-78
27	325	-155	388	-130
28	313	-168	380	-138
29	305	-185	378	-135
30	290	-170	315	-80
31	305	-173	368	-110
April 2	363	-95	368	-135
3	328	-110	360	-80
4	335	-110	373	-125
5	335	-110	370	-130
6	358	-123	305	-148
7	400	-103	398	-140
9	420	-68	423	-135
10	445	-68	433	-100
11	375	-125	308	-115
12	395	-105	378	-90
13	255	-100	395	-125
14	273	-130	430	-180
17	330	-70	433	-158
18	180	-155	240	-185
19	105	-180	373	-120
20	110	-130	275	-165
21	143	-168	383	-133
22	120	-193	393	-140
24	275	-160	448	-185
25	195	-200	395	-130
26	330	38	455	-120
27	260	-95	443	-140
28	365	85	445	-118
29	288	-85	470	-135
30	325	-45	445	-95
May 1	418	65	450	-50
2	420	-15	440	-45
3	410	115	358	-10
4	320	15	463	-78
5	430	95	450	-53
6	305	-30	443	-73
7	388	58	458	-30
8	455	125	420	5
9	373	-5	480	-45
10	460	30	380	8
11	435	-25	445	-45
12	530	53	473	-28
13	480	8	455	-20
15	513	218	463	38
16	515	110	448	15
17	523	465	448	50
18	540	160	450	20
19	568	185	420	63
20	568	340	430	33
21	525	225	418	125
22	500	235	355	178
23	490	145	410	110
24	503	225	420	183
25	480	205	448	200
26	495	240	430	250
29	510	265	423	350

Table 7. Recovery of the added NH_4^+-N and averaged inorganic N concentration in the effluent from the overland flow test models with 4% slope.

Treatment	N input		N recovered in the effluent			% of the added N removed
	conc	vol	avg vol	NH_4^+	$NO_3^- + NO_2^-$	
	µg N/ml	liter	%	---- µg N/ml	-----	
			Wastewater contains C source			
A	25.1	3	69.8	0.6	0.0	97.8
B	50.5	3	69.7	6.9	0.8	89.6
C	74.6	3	72.4	16.8	3.3	80.6
D	74.6	3	68.7	10.3	1.0	87.0
E	50.5	3	52.4	9.1	2.2	88.5
			Wastewater contains no C source			
A	25.1	3	62.8	1.7	3.3	94.2
B	50.5	3	67.0	7.1	7.7	80.9
C	74.6	3	69.0	18.4	9.1	75.1
D	74.6	3	55.0	11.7	7.7	84.7
E	50.5	3	61.6	10.9	9.4	76.1

Table 8 Distribution of added $^{15}NH_4^+$-N in the effluents from the overland flow models under various treatments with 4% slope at 22°C

Time, days	Treatment 1		Treatment 2		Treatment 3		Treatment 4		Treatment 5	
	NH_4^+	$NO_3^+ + NO_2^-$	NH_4^+	$NO_3^+ + NO_2^-$	NH_4^+	$NO_3^- + NO_2^-$	NH_4^+	$NO_3^- + NO_2^-$	NH_4^+	$NO_3^- + NO_2^-$
	μg N									
	Nitrogen-15 amendment (Input)									
0	69390.0	0.0	14939.5	0.0	22183.5	0.0	22183.5	0.0	14770.7	0.0
	Nitrogen-15 in the effluent (Output)									
0	1412.1	1402.1	407.4	200.5	2654.2	439.3	520.6	321.7	1930.1	471.8
1	396.8	2243.9	286.5	855.8	996.8	762.6	771.1	473.8		
2	70.7	901.1	90.6	224.8	327.1	267.8	735.5	261.5	166.7	477.2
3	34.9	458.6	45.8	128.7	388.9	159.9	360.8	134.0		
4	15.0	289.2	20.8	151.5	141.2	193.2	158.2	65.4	47.1	198.5
7	0.0	3.4	6.3	83.9	22.5	76.9	44.4	117.2	15.7	88.5
8	1.2	1.2	11.5	3.6	22.2	23.5	48.0	4.1		
9	0.4	0.0	9.3	2.2	23.0	11.9	27.8	0.0	10.3	48.3
10	3.1	0.0	3.9	2.8	16.3	18.6	63.4	5.2		
11	0.0	0.0	3.5	1.4	13.4	6.8	44.9	3.8	9.0	1.2
14	0.0	0.0	1.1	0.0	7.5	0.2	16.3	18.3	1.9	0.0
15	0.0	0.8	1.4	1.3	6.5	0.2	13.7	0.0		
16	0.0	0.0	1.5	0.5	4.3	0.7	8.7	0.6	1.5	18.7
17	0.5	0.0	2.2	0.4	5.4	1.1	20.3	1.7		
18	0.9	0.0	1.6	0.9	4.5	5.1	15.5	0.6	1.6	3.5
21	0.0	0.0	0.8	1.6	1.8	8.1	7.3	32.5	1.3	0.2
22	0.1	0.0	1.0	0.2	2.1	2.7	11.2	0.1		
23	0.3	0.0	1.1	0.6	1.3	1.2	7.5	0.3	0.9	0.9
24	0.0	0.0	0.6	0.4	0.3	0.9	5.8	0.7		
25	0.5	0.0	0.7	0.1	1.0	1.0	10.0	0.5	0.4	0.4
28	0.0	0.0	0.2	0.1	0.7	1.9	2.7	0.3	0.0	0.0
29	0.7	0.0	0.5	0.2	0.2	2.2	3.0	0.1		
30	1.1	0.0	0.7	0.1	0.4	1.3	5.0	0.1	0.0	0.0
31	1.0	0.0	0.7	0.1	0.0	1.3	7.2	0.2		
32	—	—	—	—	—	—	—	—	0.8	0.0
Sub Total	1939.5	5300.4	899.8	1661.9	4641.8	1988.2	2903.9	1442.7	2187.2	1309.4
TOTAL	7239.9		2561.6		6630.0		4346.6		3964.6	
% of recovery	10.43		17.15		29.89		19.59		23.67	

Table 9. Distribution of the added NH_4^+-^{15}N in various soil fractions of an overland flow system determined at the end of a 32-day investigation at 22°C (Treatment A).

Soil depth cm	Distance downslope, cm				
	0-30	30-60	60-90	90-120	overall
			NH_4^+-^{15}N (μg)		
0-3	21.81	7.83	6.64	5.86	42.14
3-6	7.47	6.60	5.35	7.30	26.72
6-10	5.93	7.74	6.96	5.61	26.24
TOTAL	35.21	22.18	18.95	18.77	95.11
			$(NO_3^- + NO_2^-)$-N (μg)		
0-3	23.62	5.79	4.23	4.36	38.00
3-6	12.45	4.07	4.15	5.87	26.54
6-10	11.84	10.19	4.86	6.40	33.29
TOTAL	47.91	20.04	13.23	16.63	97.81
			Organic ^{15}N (μg)		
0-3	2318.58	360.75	417.24	450.89	3547.46
3-6	419.52	294.85	326.68	360.25	1401.30
6-10	409.32	347.13	458.08	475.82	1690.35
TOTAL	3147.42	1002.73	1202.00	1286.96	6639.11

Table 10. Distribution of the added NH_4^+-^{15}N in various soil fractions of an overland flow system determined at the end of a 32-day investigation at 22°C (Treatment B).

Soil depth cm	Distance downslope, cm				
	0-30	30-60	60-90	90-120	overall
			NH_4^+-^{15}N (μg)		
0-3	3.75	2.37	1.49	1.43	9.04
3-6	1.50	2.00	1.81	2.04	7.35
6-10	2.52	2.07	2.65	2.16	9.40
TOTAL	7.77	6.44	5.95	5.62	25.78
			(NO_3^- + NO_2^-)-N (μg)		
0-3	7.30	1.96	1.63	1.59	12.48
3-6	3.55	1.03	0.56	0.70	5.84
6-10	3.28	1.50	0.98	1.10	6.86
TOTAL	14.13	4.49	3.16	3.39	25.17
			Organic-^{15}N (μg)		
0-3	241.69	113.52	101.34	111.10	567.65
3-6	84.82	27.90	13.66	16.96	143.34
6-10	46.44	19.76	27.53	2.38	96.11
TOTAL	372.95	161.18	142.53	130.45	807.11

Table 11. Distribution of the added NH_4^+-^{15}N in various soil fractions of an overland flow system determined at the end of a 32-day investigation at 22°C (Treatment C).

Soil depth cm	Distance downslope, cm				
	0-30	30-60	60-90	90-120	overall
			NH_4^+-^{15}N (μg)		
0-3	5.48	1.48	1.07	1.89	9.92
3-6	2.67	1.43	1.12	1.51	6.73
6-10	2.57	3.26	1.82	1.85	9.50
TOTAL	10.72	6.16	4.01	5.25	26.14
			$(NO_3^- + NO_2^-)$-N (μg)		
0-3	3.69	2.80	2.11	3.12	11.72
3-6	5.36	1.69	3.13	3.51	13.69
6-10	2.98	3.74	2.78	5.23	14.73
TOTAL	12.03	8.24	8.01	11.86	40.14
			Organic-^{15}N (μg)		
0-3	228.12	49.93	115.80	38.37	432.22
3-6	83.03	26.46	56.67	46.01	212.17
6-10	118.78	56.40	40.61	72.07	287.86
TOTAL	429.92	132.79	213.08	156.45	932.24

Table 12. Distribution of the added NH_4^+-^{15}N in various soil fractions of an overland flow system determined at the end of a 32-day investigation at 22°C (Treatment D).

Soil depth cm	Distance downslope, cm				
	0-30	30-60	60-90	90-120	overall
			NH_4^+-^{15}N (μg)		
0-3	80.21	5.81	5.21	2.84	94.07
3-6	15.34	16.89	20.03	2.75	55.01
6-10	7.40	6.47	18.24	7.58	39.69
TOTAL	102.95	29.17	43.48	13.17	188.77
			(NO_3^- + NO_2^-)-N (μg)		
0-3	20.89	13.88	1.24	5.80	41.81
3-6	45.82	46.18	5.38	16.40	113.78
6-10	66.73	29.41	58.66	55.34	209.64
TOTAL	132.89	89.47	65.28	77.54	365.18
			Organic-^{15}N (μg)		
0-3	304.45	69.46	81.26	173.43	628.60
3-6	345.21	223.18	52.28	61.94	682.61
6-10	321.38	233.33	230.07	173.29	958.07
TOTAL	971.03	525.97	363.61	408.66	2269.27

Table 13. Distribution of the added NH_4^+-^{15}N in various soil fractions of an overland flow system determined at the end of a 32-day investigation at 22°C (Treatment E).

Soil depth cm	Distance downslope, cm				
	0-30	30-60	60-90	90-120	overall
			NH_4^+-^{15}N (μg)		
0-3	2.76	2.08	1.60	2.57	9.01
3-6	1.73	2.87	2.72	3.11	10.43
6-10	2.47	4.39	1.55	1.79	10.20
TOTAL	6.95	9.34	5.87	7.45	29.61
			$(NO_3^- + NO_2^-)$-N (μg)		
0-3	8.59	3.13	2.24	4.29	18.25
3-6	4.29	1.47	1.34	1.54	8.64
6-10	3.94	2.02	1.13	1.64	8.73
TOTAL	16.82	6.62	4.72	7.48	35.62
			Organic-^{15}N (μg)		
0-3	254.06	23.85	4.06	135.47	417.44
3-6	21.53	92.89	0.00	19.74	134.16
6-10	41.56	11.98	35.50	14.45	103.49
TOTAL	317.15	128.72	39.56	169.66	665.09

Table 14. Distribution of added NH_4^+-^{15}N in ryegrass in a simulated overland flow system.

Treatment	1st harvest		2nd harvest		Total ^{15}N
	TKN	Ex ^{15}N	TKN	Ex ^{15}N	recovered
	% of dried wt	mg	% of dried wt	mg	%
A	3.63	4.259	2.39	8.444	18.31
B	3.83	1.942	2.81	1.321	21.84
C	3.71	1.477	2.60	1.056	11.42
D	3.72	1.682	2.71	1.299	13.50
E	3.71	1.519	2.59	1.325	18.20

Table 15. Distribution of added NH_4^+-^{15}N in ryegrass in various downslope distance of an overland flow system.

Treatment	Distance downslope, cm: 0-40			40-80			80-120		
	TKN	Ex A%	% Recovered	TKN	Ex A%	% Recovered	TKN	Ex A%	% Recovered
A	2.55	1.210	5.33	2.39	0.781	3.98	2.24	0.883	2.86
B	3.09	0.152	3.86	2.65	0.101	2.88	2.70	0.108	2.10
C	2.53	0.131	1.69	2.65	0.093	1.60	2.61	0.117	1.47
D	2.65	0.136	1.87	2.85	0.140	2.14	2.52	0.161	1.92
E	2.61	0.176	3.46	2.68	0.093	2.35	2.49	0.118	2.11

Table 16. Nitrogen-15 mass balance in a ryegrass-mhoon soil test model after an overland flow practice of 32 days.

	Organic N		Exch + Soluble NH_4^+-N		$(NO_3^- + NO_2^-)$-N		Total ^{15}N recovery
	μg	% of ^{15}N added	μg	% of ^{15}N added	μg	% of ^{15}N added	%
Effluent	—	—	1939.5	2.8	5300.4	7.6	10.4
Grass*	12702.7	18.3	—	—	—	—	18.3
Soil	6643.1	9.6	95.1	0.1	97.8	0.1	9.8
Total recovery	19302.5	27.9	1977.9	2.9	5398.2	7.8	38.6

*data present here including organic N and NH_4^+-N

N level in wastewater = 25 μg N/ml

Initial ^{15}N input = 6930 μg

—: Not determined

Table 17. Nitrogen-15 mass balance in a ryegrass-mhoon soil test model after an overland flow practice of 40 days.

	Organic N		Exch + Soluble NH_4^+-N		$(NO_3^- + NO_2^-)$-N		Total ^{15}N recovery
	μg	% of ^{15}N added	μg	% of ^{15}N added	μg	% of ^{15}N added	%
Effluent	—	—	899.8	6.0	1661.9	11.1	17.1
Grass*	3263.0	21.8	—	—	—	—	21.8
Soil	807.1	5.4	25.8	0.2	25.2	0.2	5.0
Total recovery	4070.1	27.2	907.3	6.2	1687.1	11.3	44.7

*data present here including organic N and NH_4^+-N

N level in wastewater = 50 μg N/ml

Initial ^{15}N input = 14939 μg

—: Not determined

Table 18. Nitrogen-15 mass balance in a ryegrass-mhoon soil test model after an overland flow practice of 40 days.

	Organic N		Exch + Soluble NH_4^+-N		$(NO_3^- + NO_2^-)$-N		Total ^{15}N recovery
	μg	% of ^{15}N added	μg	% of ^{15}N added	μg	% of ^{15}N added	%
Effluent	—	—	4641.8	20.9	1988.2	9.0	29.9
Grass*	2532.7	11.4	—	—	—	—	11.4
Soil	932.2	4.2	26.1	0.1	40.1	0.2	4.5
Total recovery	3464.9	15.6	4667.9	21.0	2028.3	9.1	45.7

*data present here including organic N and NH_4^+-N

N level in wastewater = 75 μg N/ml

Initial ^{15}N input = 22183 μg

—: Not determined

Table 19. Nitrogen-15 mass balance in a ryegrass-Rhoon soil test model after an overland flow practice of 40 days.

	Organic N		Exch + Soluble NH_4^+-N		$(NO_3^- + NO_2^-)$-N		Total ^{15}N recovery
	μg	% of ^{15}N added	μg	% of ^{15}N added	μg	% of ^{15}N added	%
Effluent	—	—	2903.9	13.1	1442.7	6.5	19.6
Grass*	2994.0	13.5	—	—	—	—	13.5
Soil	2269.3	10.2	188.8	0.9	365.2	1.6	22.2
Total recovery	5263.3	23.7	3092.7	13.9	1807.9	8.2	45.8

*data present here including organic N and NH_4^+-N

N level in wastewater = 75 μg N/ml

Initial ^{15}N input = 22183 μg

—: Not determined

Table 20. Nitrogen-15 mass balance in a ryegrass-Rhoon soil test model after an overland flow practice of 40 days.

	Organic N		Exch + Soluble NH_4^+-N		$(NO_3^- + NO_2^-)$-N		Total ^{15}N recovery
	μg	% of ^{15}N added	μg	% of ^{15}N added	μg	% of ^{15}N added	%
Effluent	—	—	2187.2	14.8	1309.4	8.9	23.7
Grass*	2688.5	18.2	—	—	—	—	18.2
Soil	665.7	4.5	29.6	0.2	35.6	0.2	14.8
Total recovery	3354.2	22.7	2216.8	15.0	1346.0	9.1	46.8

*data present here including organic N and NH_4^+-N

N level in wastewater = 50 μg N/ml

Initial ^{15}N input = 14771 μg

—: Not determined

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