In Situ Denitrification of Septic-System Nitrate Using Reactive Porous Media Barriers: Field Trials

by W. D. Robertson and J. A. Cherry[a]

Abstract

A new alternative septic-system design is presented utilizing reactive porous media barriers for passive in situ attenuation of NO_3^-. The reactive material consists of solid organic carbon (sawdust) which promotes NO_3^- attenuation by heterotrophic denitrification. Four field trials are discussed demonstrating two barrier configurations: as a horizontal layer positioned in the vadose zone below a conventional septic-system infiltration bed and as a vertical wall intercepting a horizontally flowing downgradient plume. During one year of operation both barrier configurations have been successful in substantial attenuation (60 to 100%) of input NO_3^- levels of up to 125 mg/l as N. The horizontal layer configuration can be readily installed during the construction of new infiltration beds, whereas the vertical wall configuration may be more appropriate for retrofitting existing septic systems where NO_3^- contamination has already occurred. The layer configuration allows the flexibility of constructing the barrier in the vadose zone by using coarse silt or fine sand matrix material that has the ability to remain tension-saturated, and thus anaerobic, even when positioned above the water table.

Advantages of the barrier system are that it is simple to construct, no surface structures or additional plumbing are necessary, and treatment is passive requiring no energy consumption and little or no maintenance. Mass balance calculations and preliminary results suggest that conveniently sized barriers have the potential to last for decades without replenishment of the reactive material.

Introduction

Nitrate generated by oxidation of septic-system effluent can occur in septic-system plumes at concentrations several times higher than the common drinking water limit of 10 mg/l as N (Walker et al., 1973; Robertson et al., 1991; Harman, 1992). In the ground-water zone, NO_3^- is mobile and can be unaffected by biodegradation reactions in plume zones that remain aerobic (Robertson et al., 1991). In addition, tracer tests have now demonstrated that hydrodynamic dispersion in most sand aquifers is much less than previously thought (Sudicky et al., 1983; Freyberg, 1986; Moltyaner and Killey, 1988a, b; Garabedian and LeBlanc, 1991). Thus, dilution models commonly used to attenuate NO_3^- from septic systems are probably physically unrealistic. This has been confirmed by several very detailed field studies in which above-the-drinking-water-limit NO_3^- plumes have been found to extend more than 100 m from even smaller septic systems (Robertson et al., 1991; Harman, 1992). As a result, in some jurisdictions such as the Province of Ontario, NO_3^- has become the septic-system contaminant of concern with regard to degradation of drinking water supplies.

To address the NO_3^- problem, several alternative septic-system designs for enhanced N attenuation have been investigated in recent years. The "peat" system (Brooks et al., 1984) utilizes a layer of sphagnum peat moss placed below the weeping tile bed to provide an environment for N attenuation by assimilation into fungal biomass. The "Ruuk" system (Laak, 1981) utilizes dedicated household plumbing to selectively collect toilet effluent (black water) which is nitrified and then mixed with household gray water which provides an additional organic carbon source for denitrification. A somewhat similar system is the recirculating sand filter (e.g., Piluk and Hao, 1989) in which effluent is nitrified in a sand filter and then a portion is returned to the anaerobic septic tank where carbon is available for denitrification. The latter two systems provide only partial NO_3^- attenuation, however (generally 40-90%), unless an additional carbon source such as liquid methanol is continually dosed to the final treated effluent (e.g., Sikora and Keeney, 1976; Sikora et al., 1977; Andreoli et al., 1979). An overview of existing technologies for onsite nitrogen removal is provided by Whitmyer et al. (1985).

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Although alternative systems such as these have been available for more than 10 years, none has achieved widespread usage in North America. Apparently this is because, until recently, the evidence for the occurrence of large-scale contaminant plumes from septic systems was not well-documented, with the result that regulators did not discourage the use of conventional septic systems. Thus, there has been little incentive to use the more expensive alternative designs and to accumulate the field performance data necessary to allow accurate assessment of their effectiveness.

Although NO_3^- is mobile and persistent in aerobic ground-water environments, it has long been known that NO_3^- can be attenuated in anaerobic ground-water zones when a labile carbon source is available (Trudell et al., 1986; Starr and Gillham, 1989). Attenuation is presumed to occur by heterotrophic denitrification (i.e., Delwiche, 1981):

$$5CH_2O + 4NO_3^- \rightarrow 5CO_2 + 2N_2 + 3H_2O + 4OH^- \quad (1)$$

whereby NO_3^- is biodegraded to N_2 gas. Attenuation of agriculturally derived NO_3^- in riparian ground-water zones has been attributed to denitrification resulting from increased availability of labile organic carbon in organic matter enriched sediments (Schipper et al., 1990). Likewise, Robertson et al. (1991) and Robertson and Cherry (1992) report abrupt and complete attenuation of septic-system NO_3^- in two aquifer zones where content of solid phase organic carbon in the sediments is enriched.

This paper presents a new alternative septic-system design for enhanced NO_3^- attenuation. It derives from the previous field observations of denitrification and relies on engineered anaerobic porous media barriers containing NO_3^--reactive solid organic carbon. Although in situ use of solid organic carbon has been suggested previously for denitrification of NO_3^--contaminated drinking water supplies (Boussaid et al., 1988), and in mechanically more complex, aerated, septic-system treatment schemes (Wakatsuki et al., 1993), use of reactive porous media barriers has not been suggested previously for passive in situ denitrification of septic-system NO_3^-. Although peat systems also provide passive in situ attenuation of septic-system nitrogen, the attenuation process is much different from that of the proposed reactive barrier system. Attenuation occurs by N assimilation into fungal biomass and requires aerobic conditions, whereas in the barrier system attenuation occurs by NO_3^- denitrification and requires anaerobic conditions. The N reaction products are distinctly different: organic bound N in the peat system versus N_2 gas [equation (1)] for the barrier system.

Advantages of the proposed system are that the barriers are installed in the subsurface underneath or adjacent to conventional tile beds, thus requiring no additional surface structures or plumbing, and treatment is passive so that once installed, no energy use or maintenance is required for a long period of time.

System Description

The two requirements for NO_3^- attenuation by equation (1) are the presence of an anaerobic environment and a supply of labile organic carbon to act as electron donor for heterotrophic denitrification. The denitrification barrier provides these conditions by being saturated, thus excluding atmposheric O_2 and by incorporating the necessary organic carbon as solid phase material (e.g., sawdust). The barrier can be constructed in two configurations: as a subsurface layer installed below the weeping tile field at the time of construction, or it can be retrofitted to existing septic systems as a vertical wall intercepting the NO_3^- plume at a downgradient location.

The denitrification layer is installed at a sufficient depth below the septic-system weeping tiles so that effluent NH_4^+ is oxidized to NO_3^- during percolation through the unsaturated sediments lying above the barrier layer. The carbon-augmented denitrification layer can be positioned at the water table below the tiles, or it can be positioned above the water table if the porous media material is appropriately sized to allow tension saturation of the barrier even when positioned above the water table. Coarse silt and fine sand sediments can have a hydraulic conductivity (K) in the range of 10^{-4} to 10^{-3} cm/s which is sufficient to transmit hydraulic loading of about 10 to 100 cm/day under unit hydraulic gradient. These amounts are considerably in excess of most tile bed loading rates. Yet material of this grain size also has the ability to remain saturated even when subjected to high negative pressure heads. This results from the substantial air entry value (AEV), generally 1 to 2 m, possessed by material such as coarse silt. Layers of coarse silt thus have the ability to remain tension-saturated even when positioned substantially above the water table. At many locations the position of the water table is not precisely known or it is found to be seasonally transient; thus the ability to engineer a layer of saturation at a convenient depth position below a tile field is of considerable practical importance to this technology. The preferred design for the denitrification layer is thus utilizing a porous media matrix material of coarse silt or fine sand.

Denitrification is achieved by installing an engineered sequence of porous media layers into an excavation located below the tile bed. The denitrification layer is placed at the bottom of the excavation. It consists of silty matrix material, into which solid organic carbon (e.g., sawdust) is mixed with the aid of a garden rototiller, or for larger systems, with the use of farm implements. A nitrification layer constructed of coarser grained sand is then placed above the denitrification layer.

The second configuration, the denitrification wall, can be used to retrofit existing septic systems by placement at a downgradient location if the NO_3^- plume is migrating horizontally at a shallow depth. The wall is installed below the water table perpendicular to the plume flowpath. The reactive material is placed into an excavation constructed with the aid of dewatering wells or using sheet pile. Because the barrier is placed below the water table and is not intended to impede ground-water flow, sand is the preferred matrix material for the vertical wall configuration. In selecting the porous media material, care must be excercised to ensure that permeability remains equivalent to or greater than the native sediments, or flowpath deflection around the barrier may result.

The remainder of this paper presents the results of four field trials which demonstrate several aspects of the denitrification barrier and provide performance data over about one year of operation.

These include three experiments in which the barrier was constructed as a sublayer positioned below tile fields (Long Point, Killarney, and Borden layers) and a fourth where it was installed as a vertical wall (Long Point wall).

Concurrent with the field trials, laboratory column experiments were undertaken which demonstrated the reactivity of a variety of solid carbon materials (cellulose, wheat straw, alfalfa straw, and sawdust) in promoting passive denitrification in porous media barriers (Vogan, 1993).

Long Point Silt Layer

Use of selective porous media layering to maintain saturated zones above the water table has been considered previously as a possible method for reducing atmospheric oxygen diffusion and resulting acid generation in reactive mine tailings (Nicholson et al., 1989). Because gas diffusivity through porous media decreases by more than three orders of magnitude, as degree of saturation increases (Reardon and Moddle, 1985), oxygen flux from the atmosphere to the subsurface is greatly reduced when a zone of saturation is encountered. The concept of using fine over coarse layers to maintain saturation within the overlying fine layer was explored in detail by Nicholson et al. (1989). This was followed by numerical simulation of the drainage of layered systems (Akindunni et al., 1991) and laboratory column studies utilizing layers of fine over coarse sand (Nicholson et al., 1991) and silt till over sand (Yanful, 1991). From these studies it was recognized that maintenance of saturation in the fine layer, during prolonged drainage, was dependent upon development of a condition of "static" hydraulic non-equilibrium, reached at a point when the moisture content of the underlying coarse layer approached residual saturation. Further drainage of the overlying fine layer was then impeded by the exceedingly low hydraulic conductivity of the drained, coarse layer. Under such conditions the negative pressure head at the fine/coarse interface is maintained at a constant value corresponding to the value at which the coarse layer reaches residual saturation, and is independent of the position of the water table. The thickness of the fine layer that can then remain saturated is equal to the air entry value of the fine layer minus the magnitude of the pressure head at which the coarse layer reaches residual saturation (Nicholson et al., 1990).

In this field trial a silt layer, 64 m^2 in area, was installed in the sandy vadose zone below the Long Point infiltration bed which receives effluent from a seasonal use campground. Vadose zone moisture content and pore gas O_2 levels were then monitored for a one year period after installation, including an episode of prolonged drainage during which no effluent was discharged to the tile field. The hydrogeology of the site and the hydrochemistry of a ground-water plume emanating from an older infiltration bed at the site is described by Robertson and Cherry (1992).

Methodology

In June 1990, approximately one-quarter of the new tile bed was excavated to a depth of 1.5 m, and a 25 cm thick silt layer was installed in the bottom of the excavation over an 8 × 8 m area (Figure 1). Excavation and silt placement was done using a backhoe. After placement, the silt layer was compacted while under a condition of near-saturation, then the spoil material and weeping tile lines were replaced to their original configuration. Figure 2 shows a profile through the 2 m thick vadose zone below the modified tile bed area. The silt layer is positioned at an average depth of

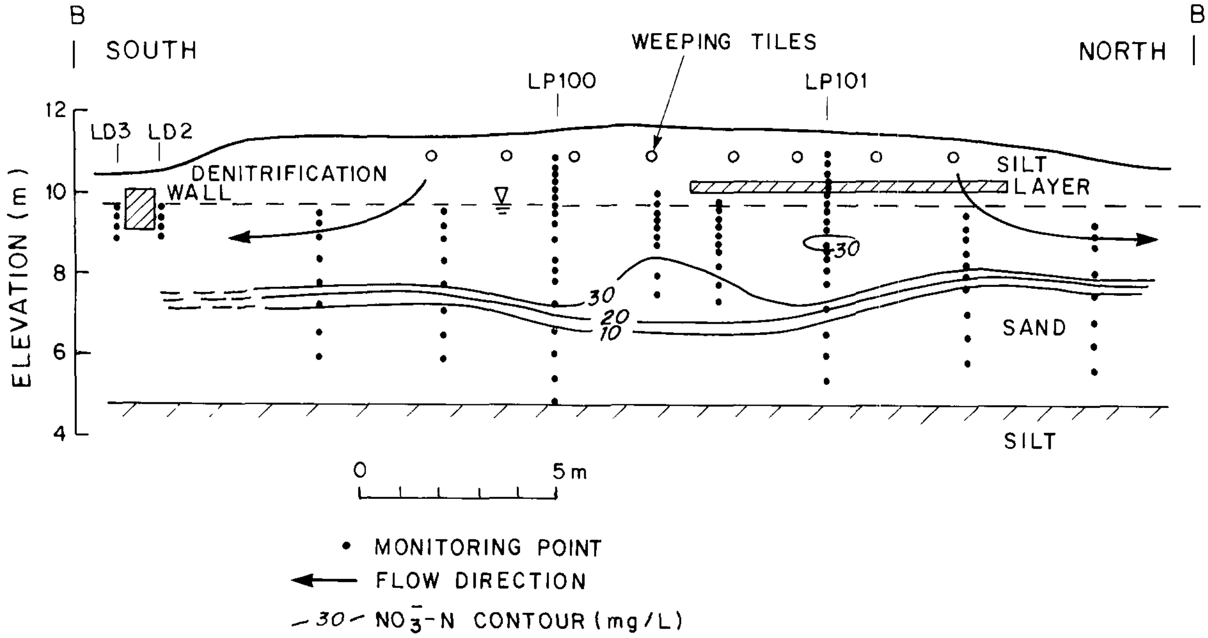

Fig. 1. Cross section through Long Point tile bed showing NO_3^--N distribution and location of engineered unsaturated zone silt layer and denitrification wall.

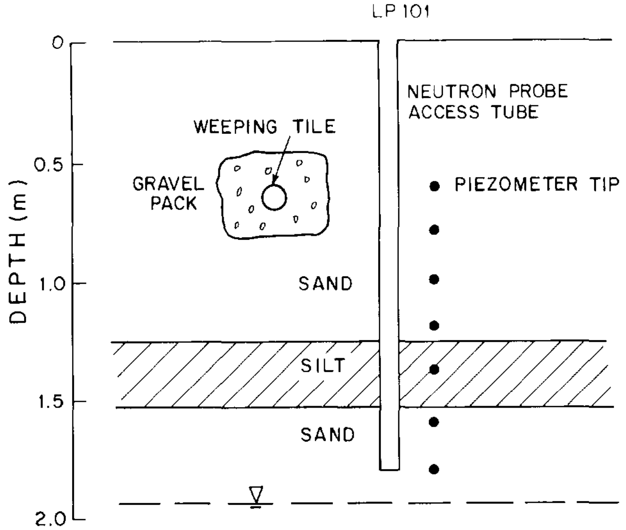

Fig. 2. Unsaturated zone profile through the modified tile bed area, Long Point, showing monitoring network installed.

1.27 to 1.52 m below ground surface which is about 0.5 m above the average water-table position. The tile lines are centered at a depth of 0.6 m and are encased by centimeter-sized gravel for a radius of about 0.15 m. After layer installation, continuous 5 cm diameter sediment cores were retrieved to a depth of 7 m, from the center of the silt bed area (core 101, Figure 1) and from a control site in the unmodified tile bed area (core 100). Cores were collected in 1.5 m long aluminum core tubes using the apparatus and methodology of Starr and Ingleton (1992). Multiple piezometer bundles were then inserted into the core holes for the purpose of ground-water sampling and to enable vadose zone gas sampling. Piezometers were constructed of 3 mm diameter polyethylene tubes with short centimeter-long screened tips. Through the vadose zone, tips were positioned at 0.2 m depth intervals (Figure 2). Adjacent to piezometers 100 and 101, permanent neutron probe access tubes of 5 cm diameter aluminum were installed to 1.8 m depth, to enable vadose zone moisture content measurements.

The entire effluent load from the campground comfort station was directed to the modified tile bed beginning in June 1990. Vadose zone moisture contents and pore gas O_2 concentrations were then measured on July 11 and August 14, 1990, during high effluent loading (11 and 9 cm/day, respectively) and on November 3, 1990 and November 26, 1991, during low loading (1 and 0.4 cm/day, respectively). Loading was calculated from the metered park water usage, except for the latter monitoring event which occurred 15 days after the park was closed for the season. Loading at this time was estimated as the average precipitation amount that occurred during the previous two weeks. Samples of vadose zone pore gas were obtained from the piezometer tubes using a peristaltic pump, then measurements of O_2 content were made, in line, using a NOVA model LBD50 field portable meter, calibrated prior to use, to the atmospheric O_2 value (20.9%). Measurements of vadose zone moisture content were made using a CPN Corp. model 503 DR neutron probe. Measurements were made at 7.5-15 cm depth increments and had an effective radius of influence of about 15 centimeters.

Sediment grain-size analyses were completed by sieve for the sand fraction and hydrometer for the clay and silt fractions. The silt layer porosity was estimated by measuring the difference in saturated and oven-dried weights of six recompacted samples.

Sediment Characteristics

Table 1 gives the grain-size properties for the silt layer material and the underlying and overlying vadose zone sand. The sand is shown to be well-sorted, silt-free, and of fine to medium texture. The sand air entry value was estimated to be about 25 cm (Table 1) based on the d_{10} grain-size diameter and the relationship given by Nicholson et al. (1991). The "silt" is in fact well-sorted, silty, very fine sand and has an estimated AEV of 130 cm. The average porosity of the six samples of recompacted silt was 0.32 ($s = 0.008$) which is typical of unconsolidated sandy deposits (Freeze and Cherry, 1979). The porous media characteristics of the two sediments of interest in this field test are not unlike those used by Akindunni et al. (1991) in simulating drainage of a similarly layered system (Touchet silt overlying medium sand).

Moisture Profiles

Figure 3 shows vadose zone moisture profiles in the center of the silt layer area (LP 101) and at the control site (LP 100) where the vadose zone consists entirely of sand. Effluent loading during the four measurement episodes ranged from 11 to 0.4 cm/day. Moisture content in the shallow vadose zone is shown to vary from about 10 percent during low loading to 15-20 percent during high loading. The lower amount probably approaches the residual saturation value for the sand. During each measurement episode, maximum moisture contents measured within the 25 cm thick silt layer ranged from 34.0 to 34.8 percent. These values slightly exceeded the estimated porosity of the silt (0.32); thus at least part of the layer was at or near saturation during each measurement. This occurred even though the water table was positioned 0.3 to 0.5 m below the bottom of the silt layer. On each occasion, except possibly during the 11 cm/day monitoring episode when the water table was higher than on the other occasions, the sand below the silt exhibited various degrees of desaturation as indicated by declining moisture contents with depth.

Oxygen Profiles

Figure 3 also shows pore gas O_2 concentrations during three of the measurement episodes. Shallow O_2 levels (0.6 m depth) varied from 10.2 percent of pore gas volume during high loading to a near-atmospheric value of 20.5 percent during low loading. On each occasion shallow O_2 levels were somewhat lower at the control site (LP 100). A possible explanation is that during installation of the silt layer, the soil zone and grass cover was disturbed causing increased O_2 diffusivity through the soil zone in that area. With increasing depth at both monitoring sites, O_2 levels became depleted. Greatest O_2 declines were noted immediately below the

Table 1. Sediment Grain-Size Distribution, Hydraulic Conductivity (K), and Air Entry Value (AEV) in the Unsaturated Zone, Long Point Silt Layer Site (Core 101)

Core Depth (m)	Material	M-C Sand >0.25 mm	F Sand >0.053 mm	Silt	Clay <0.002 mm	d_{10} (mm)	K^1 (cm/s)	AEV^2 (cm)
		(wt%)						
1.20	sand	33	65	0.4	1.2	0.15	2×10^{-2}	25
1.48	silt	0	62	37	0.9	0.02	4×10^{-4}	130
1.52	silt	2	57	41	0.4	0.02	4×10^{-4}	130
1.80	sand	22	78	0.0	0.8	0.15	2×10^{-2}	25

[1] Estimated from d_{10} grain-size diameter using Hazen formulae.

[2] Estimated from d_{10} grain-size diameter using relationship of Nicholson et al. (1991).

weeping tiles indicating that this was the zone where oxidation of effluent organic matter and NH_4^+ was most intense. Pore-water monitoring showed that on most occasions, by 1.3 m depth, the effluent NH_4^+ load averaging 66 mg/l-N, was completely oxidized to NO_3^- and the DOC load, averaging 21 mg/l, was about 75 percent depleted (Cherry and Robertson, 1992). At the control site (LP 100), minimal additional oxygen depletion was noted below about 1.2 m depth. At the silt site, however, further abrupt depletion of O_2 was consistently noted at the depth of the silt layer (1.5 m). During high loading, O_2 levels of 7.7 and 14.3 percent occurring at the top of the layer were depleted to values of 1.0 and 2.1 percent, respectively, below the layer, while during low loading, O_2 values decreased from 19.8 to 10.5 percent across the layer.

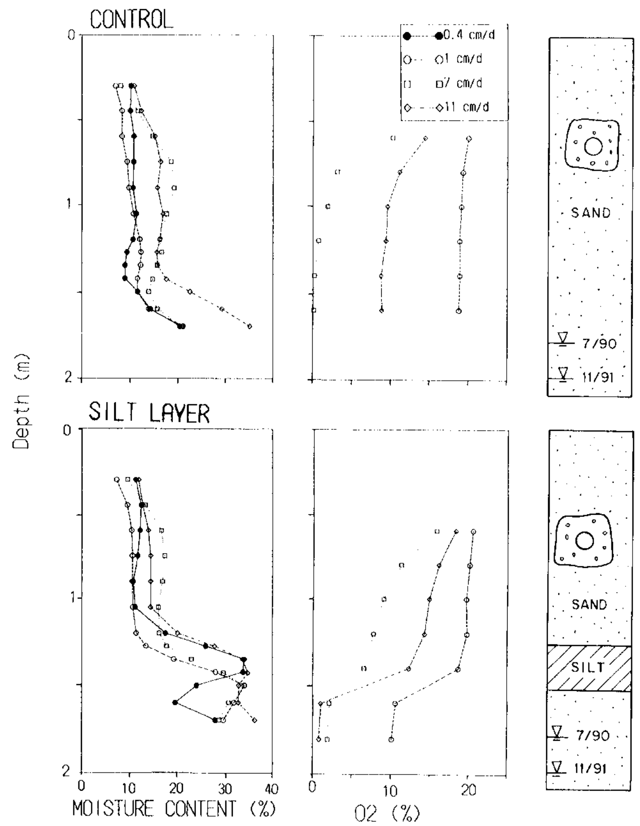

Fig. 3. Profiles of vadose zone volumetric moisture content and pore gas O_2 content below the Long Point tile bed for various rates of effluent loading: (a) control site (LP 100), and (b) silt layer site (LP 101).

The abrupt depletion of O_2 levels across the silt layer provides important evidence that such a layer, when saturated, can act as an effective O_2 diffusion barrier in a field setting. Monitoring over a wide range of recharge rates has shown that such a layer can have a rather robust capability for maintaining saturation. Of particular interest is the November 26, 1991 monitoring episode that was completed 15 days after effluent loading to the tile bed was discontinued for the season. The numerical simulations of Akindunni et al. (1991) indicate that during drainage of similarly layered porous media, conditions of "static" non-equilibrium are generally reached by about 14 days, after which little addition change in moisture content or hydraulic head occurred. Although a substantial amount of precipitation (0.4 cm/day) occurred in the two weeks prior to this monitoring event, it can be considered somewhat representative of the moisture profile to be expected when septic systems are periodically disused. The position of the water table, 50 cm below the silt layer at that time, suggests that the pressure head in the underlying sand may have approached −50 cm below the silt/sand interface. Because the silt layer is 25 cm thick, the pressure head at the top of the layer may thus have approached −75 cm, a value which remained substantially less than the estimated AEV for the silt (−130 cm). Maintenance of saturation within the silt layer was thus consistent with the conceptual view of Nicholson et al. (1989) and the simulation results of Akindunni et al. (1991). For the field configuration investigated here, that is a 25 cm thick silt layer with an estimated AEV of 130 cm, full saturation should be maintained during drainage, regardless of position of the water table, when the magnitude of the residual saturation pressure of the underlying sediment is about −100 cm or less. Most silt-free sands would reach residual saturation at pressures less than this amount (Gillham, 1984).

In June 1991, one year after the silt layer was placed, 12, one m^2 plots were excavated to the top of the layer. The sand/silt interface was found to remain distinct and planar at that time and no evidence of biomat buildup, fracturing, or macropore development was observed in the silt. Thus, it was demonstrated that the physical integrity of such a barrier layer could be maintained in a field setting when protected by an overlying layer.

Although the Long Point silt layer proved effective as an oxygen diffusion barrier, it was found to be ineffective in promoting denitrification because it was not augmented with organic carbon material.

Killarney and Borden Denitrification Layers

Two additional field trials were initiated in which silt layers similar to that at Long Point were installed in a test plot at a seasonal use family cottage located in central Ontario (Killarney site) and at a septic system servicing a seasonal use trailer camp also in central Ontario (Borden site). During these trials, however, solid phase organic carbon was added to the silt material.

A small test plot 1.95 m^2 in surface area was constructed adjacent to the existing tile field at the Killarney site and was fed by periodic pumping from the septic tank beginning in May 1992. The tank receives effluent from toilet, dishwashing, and washbasin facilities in the cottage but no showering or clothes laundering is done. The Killarney septic system is described in more detail in an accompanying paper (Robertson and Blowes, 1995).

At the Borden site, flow from the septic tank servicing the trailer camp washhouse was diverted entirely to the experimental tile field, 18 m^2 in surface area, installed in July 1992. The septic tank receives effluent from toilet, washbasin and showering facilities only.

Methods

At the Killarney site the test plot consisted of weeping tiles of perforated PVC pipe encased for a radius of 10 cm by washed gravel, overlying 30 cm of medium sand. The sand layer was in turn underlain by a 60 cm thickness of engineered silt in three distinct layers (Figure 4). The uppermost layer consisted of the same silt material used at Long Point (see Table 1) but mixed with coarse sawdust, the middle layer consisted of the native lacustrine silt spoil material from the excavation, mixed with leaf compost, while the bottom layer consisted of native silt mixed with unprocessed grain seed. Each of the silt layers contained about 15% by volume of the carbon material. Also, a 3 centimeter thick layer of crushed limestone was placed at the bottom of the sand layer to buffer acidity generated by effluent oxidation. The engineered layers were framed with plywood to assure 1-D (downward) flow of the effluent. Operation of the test plot involved periodic closing with septic tank effluent, generally in 20 liter increments, beginning in May 1992 and continuing until August 1993.

At the Borden site the experimental tile bed consisted of PVC pipe weeping tiles positioned in a semicontinuous gravel layer that was separated from the silty layer by 40 cm of medium sand (Figure 4). The 0.5 m thick silt layer is positioned at a depth of 0.7 to 1.2 m and in turn overlies a bottom layer of medium sand. The silt layer consists of the same silt material used at Long Point (Table 1), but uniformly mixed with approximately 15 vol % sawdust. The engineered layers were again framed with plywood to assure downward flow of the effluent.

Pore-water monitoring was achieved using porous cup suction lysimeters installed in the silt layers at the depths shown on Figure 4 and using PVC pipe piezometers installed in the sand layers above and below the silt layers. Sampling ports were drilled into the weeping tiles to allow sampling of the effluent. Samples were obtained from the lysimeters by applying suction, waiting several hours, and then pumping the well bore water using a peristaltic pump and polyethylene tubing. Samples were field-filtered (0.45 μm) and were accompanied by field measurement of pH, calibrated against buffers of pH 4 and 7, and measurement of electrical conductance (EC). Detailed sampling was completed on nine occasions during May 1992 to August 1993 at Killarney and on six occasions during July 1992 to September 1993 at Borden. Samples were generally analyzed for a complete suite of major ions (Cl^-, SO_4^{2-}, NO_3^-, PO_4^{3-}, Ca^{2+}, NH_4^+, Mg^{2+}, Na^+, K^+, Fe, Mn, Si), alkalinity, and dissolved organic carbon (DOC). Analyses were completed within one to two weeks of sample collection by ion chromatography and colorimetric techniques (anions, NH_4^+, and DOC) and by atomic absorption spectrometry (cations).

Layer Residence Time

At the Killarney site, the test plot was dosed with septic tank effluent in measured increments; thus, effluent loading and pore-water residency in the silt layer could be estimated directly. During the 424 day monitoring period, total effluent loading to the 1.95 m^2 test bed was 1990 liters, representing 1-D loading of 0.88 m/a. Precipitation recharge on flat-lying silty overburden terrain is likely to be on the order of 0.2 m/a in this climatic region (Robertson and Cherry, 1989); thus, total loading (effluent + precipitation) is estimated at 1.1 m/a. Assuming porosity of 0.32, residency in the 0.35 m thick silt-sawdust layer positioned at 0.4 to 0.75 m depth would average about 40 days. During initial monitoring, Cl^- at the bottom of the silt-sawdust layer increased from 11 to 17 mg/l between days 30 and 40 after startup and remained above 15 mg/l thereafter, confirming the above estimate of residency.

At the Borden site, loading to the experimental tile bed was not measured directly and was dependent upon the number of users of the trailer facilities, generally one to five during May to October, with little loading during November to April. Assuming average effluent generation of 200 liters/day during the summer season, based on two person usage (100 liters/day/person), loading would average 1.1 cm/day. Again assuming porosity of 0.32, residency in the 0.5 m thick silt-sawdust layer would be about 15 days. This estimate was confirmed by initial monitoring which showed Cl^- levels at the bottom of the silt layer, increasing from a value of 11 mg/l 12 days after startup, to 48 mg/l 20 days after startup.

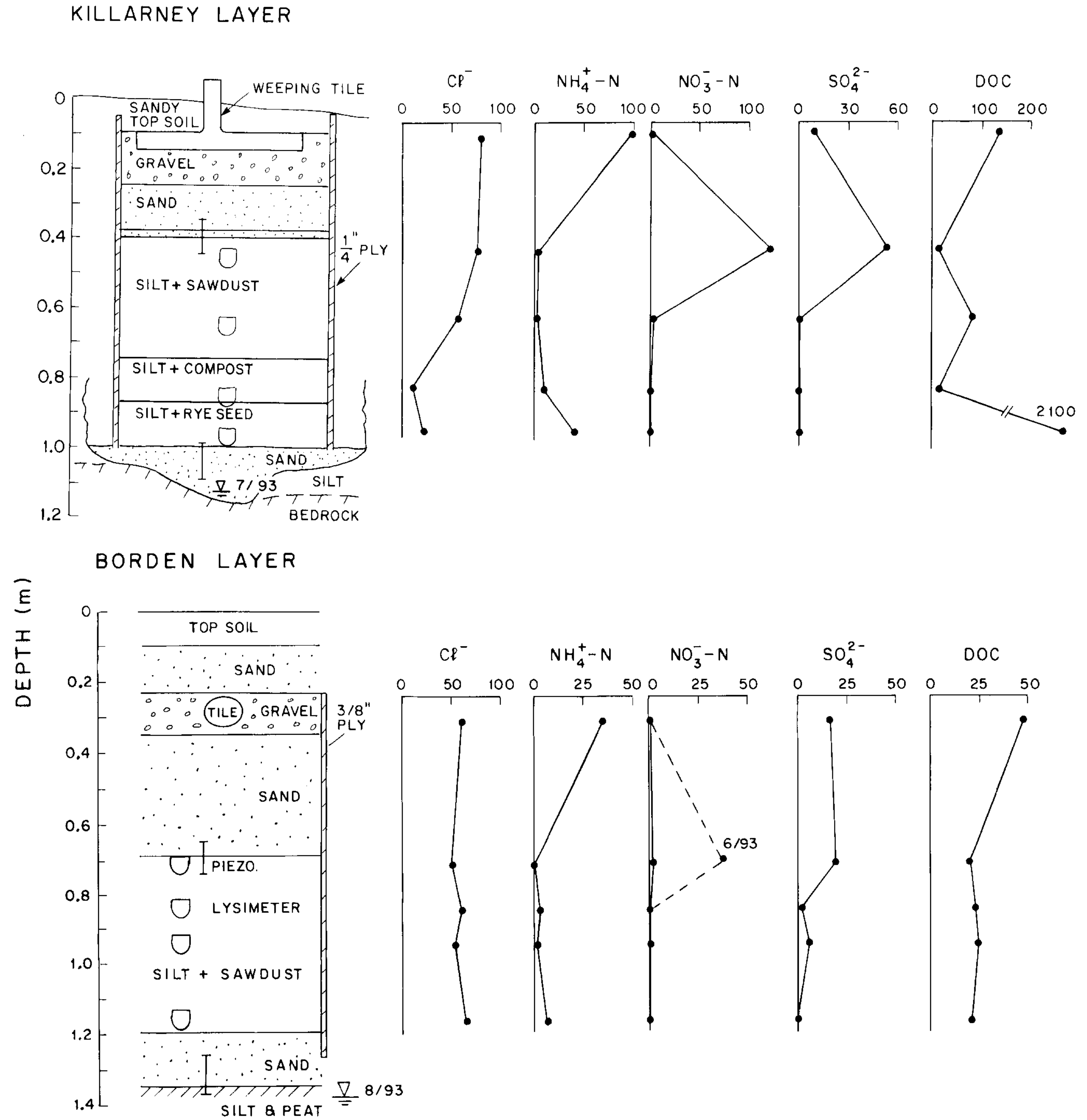

Fig. 4. Killarney and Borden denitrification layers showing chemical profiles (mg/l) after one year of operation (410-424 days after startup, Killarney site, July 16-30, 1993; 395 days after startup, Borden site, August 23, 1993).

Results

Figure 4 shows vertical profiles of Cl^-, NH_4^+, NO_3^-, SO_4^{2-}, and DOC after about one year of operation of the Killarney and Borden denitrification layers. Figure 5 shows the trends of NH_4^+, NO_3^-, and DOC breakthrough at the bottom of the silt-sawdust layers over the one year period. Tables 2 and 3 compare the major ion chemistry of the tile effluent to that of the pore water at the top and bottom of the silt-sawdust layers.

At the Killarney site, sampling on day 424 showed that effluent NH_4^+-N (99 mg/l) was almost entirely depleted by oxidation to NO_3^- in the sand layer overlying the denitrification barrier (Figure 4). The resulting NO_3^--N level of 125 mg/l occurring at the top of the barrier was then attenuated to a very low level (1.2 mg/l) at the bottom of the silt-sawdust layer, indicating almost complete denitrification. DOC levels decreased from an effluent value of 134 mg/l to a low value of 10 mg/l within the upper sand layer, further indicating substantial oxidation in this zone. DOC then increased to 86 mg/l at the bottom of the silt-sawdust layer indicating some excess leaching of the solid carbon material. Extremely excessive DOC values (> 2000 mg/l) were consistently observed in the lower silt-rye seed layer; thus, interest here is focused primarily on the shallower sawdust layer. Abrupt attenuation of SO_4^{2-} from a level of 54 mg/l to < 2 mg/l (Table 2) in the sawdust layer indicates that conditions sufficiently reducing for SO_4^{2-} reduction were also present. Lower Cl^- values occurring below the silt-sawdust layer (10-20 mg/l, Figure 4) reflect loading that occurred in the Spring of 1993 when dilution from precipitation was substantial.

After one year of operation, chemical profiles below the Borden tile field were generally similar to those observed at Killarney (Figure 4). NH_4^+ in the effluent (34 mg/l as N) was attenuated to low values (< 0.05-7.7 mg/l) in the silt-sawdust layer suggesting that significant oxidation to NO_3^- occurred in the overlying sand layer. High NO_3^- values were observed only sporadically at the top of the silt-sawdust layer, however (e.g., 37 mg/l as N, 6/93), suggesting that denitrification was frequently active even at the depth position of the shallowest lysimeter (0.7 m). Attenuation of SO_4^{2-} values from 18 mg/l at the top of the silt layer to < 2 mg/l at the bottom, demonstrates that SO_4^{2-} reduction was again also active. DOC decreased from an effluent value of 47 mg/l and then remained in range of 20-25 mg/l through the silt barrier.

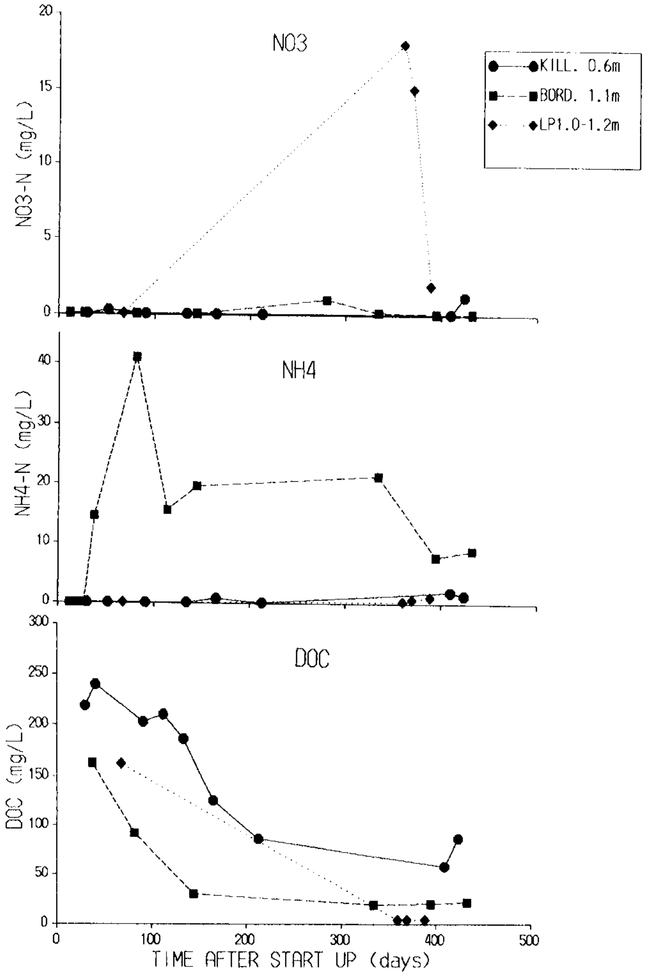

Fig. 5. Trends of NO_3^-, NH_4^+, and DOC in pore water exiting the denitrification barriers at Killarney, Borden, and Long Point.

While NH_4^+ values were generally low (< 8 mg/l as N) throughout the Borden silt layer on day 395, examination of the trend of NH_4^+ breakthrough at the bottom of the layer (Figure 5) shows that significant NH_4^+ (15-41 mg/l as N) was present in the silt pore water during earlier monitoring. This reflects less complete oxidation of effluent NH_4^+ that occurred as a result of high water-table levels experienced in the Summer and Fall of 1992 and also as a result of the presence of an excessively thick topsoil layer that likely impeded atmospheric O_2 diffusion into the subtile sand layer. After removal of the topsoil layer in April 1993 (day 280) and as a result of lower water-table levels experienced in 1993, decreased NH_4^+ levels (< 10 mg/l as N) were observed at the top of the silt layer in 1993, reflecting more complete effluent oxidation. Although some NH_4^+ (8-41 mg/l as N) is shown to persist at the bottom of both the Killarney and Borden layers after the first year of operation (Figure 4), it is considered unlikely that the NH_4^+ results from dissimilatory NO_3^- reduction. If this were the case, high NH_4^+ levels would also be present in the shallower horizons where NO_3^- attenuation is most intense. The NH_4^+ that persists is likely the result of incomplete effluent nitrification at Borden and the result of mineralization of organic N leached from the rye seed present in the lower layer at Killarney.

During monitoring in the Summer of 1993, the water-table levels at both sites were at least 10 to 20 cm below the bottom of the silt barriers. Nonetheless, vigorous and complete denitrification occurred within the top 20 cm of both barriers, demonstrating that a high degree of saturation was maintained in the barriers even though positioned substantially above the water table. Hydraulic performance was thus consistent with that of the Long Point silt layer discussed earlier.

Long Point Denitrification Wall

In September 1992 a vertical wall configuration of the denitrification barrier was installed at the Long Point site. The barrier 0.6 m × 1.2 m in surface area was installed to a depth 0.8 m below the water table (Figure 6). The barrier was positioned perpendicular to the direction of ground-water flow at a location 10 m from the edge of the tile bed, where NO_3^- contaminated shallow ground water (24-64 mg/l NO_3^--N) was migrating horizontally away from the tile bed (Figure 1). A plywood frame was used to assist in excavation and wall placement, which was then removed after barrier installation. The barrier porous media consists of the medium sand spoil material mixed with about 20% by vol. coarse sawdust. Flow velocity at this location, based on the estimated hydraulic conductivity of the sand (2×10^{-2} cm/s, Robertson and Cherry, 1992) and the observed hydraulic gradient to the south (0.002) was estimated to be about 20 m/a during the high loading summer season. Ground-water residence time within the 0.6 m thick barrier was thus estimated to be about 10 days, although this value varies depending on tile bed loading rate.

Pore-water monitoring was achieved using two nests of four piezometers each, positioned at the upgradient (LD2) and downgradient edge (LD3) of the wall (Figure 6). These were constructed of 1.3 cm diameter PVC pipe with short (5

Table 2. Chemistry of the Killarney Denitrification Layer 424 Days After Startup (July 30, 1993)

	Tile Effluent (16/7/93)	Top of Layer (0.4 m)	Bottom of Sawdust Zone (0.6 m)
Na^+ (mg/L)	58	57	7.6
K^+	36	20	7.9
Ca^{2+}	12	225	135
Mg^{2+}	6.2	40	21
Alk ($CaCO_3$ equiv.)	421[1]	257[1]	393[1]
SO_4^{2-}	9	54	<2
Cl^-	82	77	59
Si	3.8	11	45
PO_4^{3-}-P	3.5	<0.01	<0.01
NO_3^--N	0.08	125	1.2
NH_4^+-N	99	4.6	1.4
Fe	0.71	<0.02	39
Mn	0.12	2.4	6.4
DOC	134	10	86
EC (umhos/cm)	1283	1810	968
pH	6.50	6.75	6.60

[1] Measured in the laboratory.

Table 3. Chemistry of the Borden Denitrification Layer 395 Days After Startup (August 23, 1993)

	Tile Effluent	Top of Layer (0.7 m)	Bottom of Layer (1.1 m)
Na^+ (mg/L)	82	55	36
K^+	15	7.9	9.1
Ca^{2+}	65	106	102
Mg^{2+}	18	14	14
Alk ($CaCO_3$ equiv.)	416[1]	380[1]	315[1]
SO_4^{2-}	15	18	<2
Cl^-	61	50	63
Si	22	19	14
PO_4^{3-}-P	5.8	<0.01	<0.01
NO_3^--N	<0.05	0.26	<0.05
NH_4^+-N	34	<0.05	7.7
Fe	0.65	0.80	16
Mn	0.17	2.2	4.3
DOC	47	20	20
EC (umhos/cm)	1082	904	872
pH	6.80	6.65	6.40

[1] Measured in the laboratory.

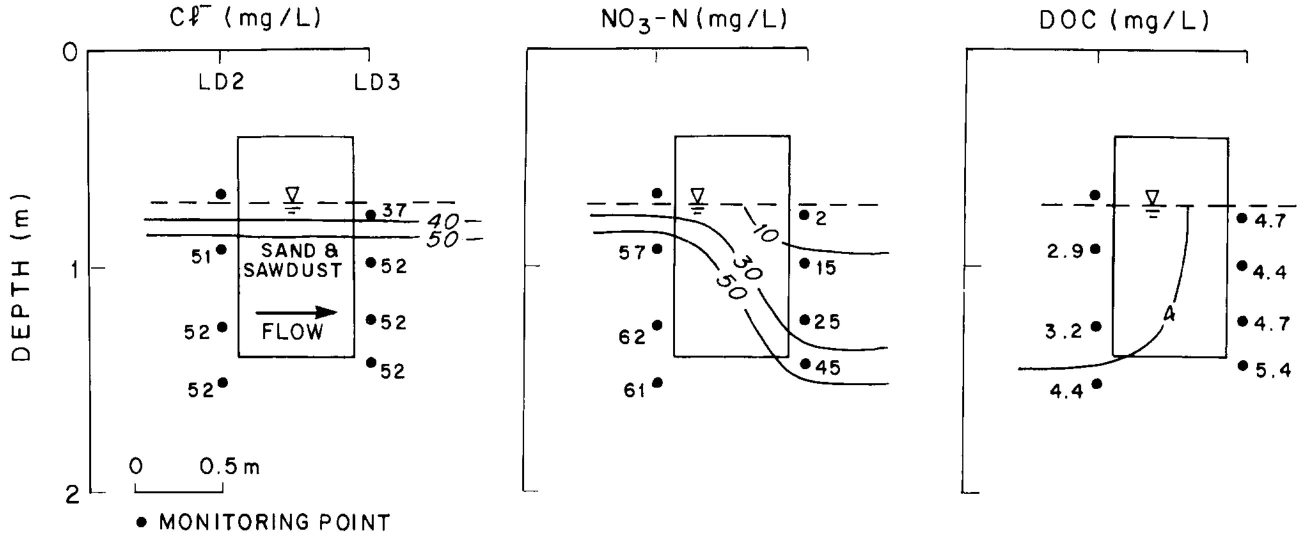

Fig. 6. Long Point denitrification wall showing Cl$^-$, NO$_3^-$-N, and DOC levels upgradient and downgradient of the wall after 370 days of operation (September 17, 1993).

cm long) slotted tips. Sampling was done using the same apparatus and techniques as at Killarney and Borden, except that in this case, sample volumes were sufficient to allow field determination of alkalinity by acid titration. Samples were obtained on nine occasions during the 370 day monitoring period.

Results

Figure 6 shows concentrations of Cl$^-$, NO$_3^-$, and DOC upgradient and downgradient of the barrier wall after about one year of operation. Table 4 compares plume-water composition upgradient and downgradient of the wall at that time. These samples were obtained on September 17, 1993, 10 days after the period of maximum summer season effluent loading. Pore-water residency in the barrier wall was estimated to be about 10 days at that time. Consistent Cl$^-$ levels (51-52 mg/l) present below 0.9 m depth demonstrate that undiluted septic plume water was migrating through the bottom half of the barrier. Very high NO$_3$-N values present upgradient of the wall (57-62 mg/l) were attenuated to much lower values (2-25 mg/l) downgradient. DOC levels at the downgradient edge of the barrier (4.4-4.7 mg/l) were only slightly elevated compared to upgradient values (2.9-3.2 mg/l). Generally, similar results were observed during earlier monitoring at high to medium loading periods, except that higher DOC levels occurred downgradient, and were accompanied by more complete NO$_3^-$ attenuation (Figure 5), and also significant SO$_4^{2-}$ attenuation occurred, again indicating SO$_4^{2-}$ reduction.

Sensitivity of the barriers' effectiveness to pore-water residence time was demonstrated by the behavior of NO$_3^-$ during days 360 to 389. Day 360 occurred on September 7, 1993, the day after the end of peak summer season effluent loading. At this time water-table mounding and horizontal ground-water velocities near the tile bed were expected to be at a maximum. NO$_3^-$ treatment efficiency was observed to be 72% at this time (64.5 mg/l NO$_3^-$-N upgradient, versus 18.0 mg/l downgradient). Treatment efficiency then improved over the next 29 days as loading to the tile bed diminished, resulting in lower ground-water velocities and increased barrier residence time. On September 17 treatment efficiency improved to 76% (61.5 mg/l NO$_3^-$-N upgradient, versus 15.0 mg/l downgradient), while by October 6 treatment improved substantially to 97% (63.5 mg/l upgradient, versus 1.9 mg/l downgradient). Park water usage in September (100 m^3/week) averages about one-third of the peak season amount (300 m^3/week); however, the hydraulic gradient in the area near the wall (about 0.002) was too small to allow accurate observation of changing gradient during this period.

Discussion

Barrier Longevity

These field trials have demonstrated the feasibility of using reactive porous media barriers to achieve in situ denitrification of septic-system NO$_3^-$. However, in many situations, such barriers would be considered useful only if they remained effective for long periods without maintenance. Thus, longevity of the organic carbon material is crucial to this technology. If it is assumed that the barrier organic carbon is consumed only by heterotrophic denitrification [equation (1)], then it is easy to demonstrate that a denitrification layer of modest thickness has the potential to last a very long time. Consider a typical single family residence generating 1000 liters/day of effluent with average NH$_4^+$-N content of 40 mg/l. Potential annual NO$_3^-$-N loading would thus be 15 kg. A one meter thick barrier layer, 100 m^2 in area and containing 2 wt % organic carbon, would contain 3600 kg of organic carbon. Because denitrification of 1 kg of NO$_3^-$-N by equation (1) requires about 1 kg of C, the carbon mass would be sufficient for 200 years of denitrification. Even if only 10% of the organic carbon was available for denitrification, the layer would still last for the typical design life of a septic system, about 20 years.

Table 4. Chemistry Upgradient and Downgradient of Long Point Wall 370 Days After Startup (September 17, 1993)

	Upgradient (LD2-1.3 m)	Downgradient (LD3-1.3 m)
Na^+ (mg/L)	34	27
K^+	7.2	5.7
Ca^{2+}	192	173
Mg^{2+}	22	20
Alk ($CaCO_3$ equiv.)	370[1]	390[1]
SO_4^{2-}	18	27
Cl^-	52	52
Si	10	10
PO_4^{3-}-P	<0.01	<0.01
NO_3^--N	62	25
NH_4^+-N	<0.05	0.05
Fe	0.20	4.2
Mn	1.1	1.7
DOC	3.2	4.7
EC (umhos/cm)	1433	1183
pH	6.85[2]	6.55[2]

[1] Measured in the field.
[2] Measured September 10, 1993.

Experience at the Killarney, Borden, and Long Point sites suggests that during initial operation at least, barrier carbon loss will also result from SO_4^{2-} reduction and excess DOC leaching. Consider excess DOC leached from the bottom of the Borden layer. DOC levels were initially about 150 mg/l, but then declined to a uniform value of about 20 mg/l after 100 days of operation (Figure 5). Using the previous estimate of average effluent loading (200 liters/day) during the six-month seasonal usage period, annual loading would be 36,000 liters. Assuming the DOC value of 20 mg/l is entirely carbon derived from the barrier, annual carbon loss by excess leaching would be 0.7 kg. It is estimated that 150 kg of carbon is present in the Borden barrier layer, thus < 1% would be lost by excess leaching annually. Carbon consumption from SO_4^{2-} reduction and dissolved O_2 reduction would be less than that from denitrification because of the lower molar quantities of SO_4^{2-} (0.19-0.56 mmol/l, from Tables 2-4) and dissolved O_2 present (maximum of 0.5 mmol/l) compared to $NH_4^+ + NO_3^-$ (2.4-9.2 mmol/l, from Tables 2-4).

Thus, such calculations suggest that conveniently sized barriers have the potential to provide effective denitrification for a considerable number of years without maintenance.

Side Effects

Changes in barrier water quality that might be considered detrimental are the development of increased levels of Fe, and at times DOC. After one year of operation, discharge water from the three barriers had Fe that varied 4.2 to 39 mg/l and DOC that varied from 3.7 to 86 (Tables 2-4) with concentrations correlated to pore-water residence time in the barriers.

Iron is presumed to derive from Fe^{2+} that is librated during reductive dissolution of Fe oxyhydroxide minerals present in the porous media. Fe^{2+} can be relatively mobile in flow systems and can lead to generation of acidity when oxidized to Fe^{3+} at surface discharge points. However, because of elevated alkalinity levels in these waters (315 to 393 mg/l, Tables 2-4) decrease in pH from iron oxidation would be minor. Concerns about Fe may result primarily from aesthetic problems associated with precipitation of Fe oxyhydroxide minerals upon atmospheric exposure and related effects such as clogging of well screens. It is possible that other trace metals may also be released when Fe is mobilized.

In some environments, however, such as near sensitive aquatic ecosystems, increased Fe levels might be considered of benefit because of the potential to limit PO_4^{3-} concentrations by precipitation of vivianite. Although PO_4^{3-}-P levels are low at these barrier locations (<0.01 mg/l), possibly as a result of PO_4^{3-} sorption reactions occurring closer to the tile lines, it is likely that PO_4^{3-} will migrate slowly in these flow systems as sorption capacity is consumed. Because PO_4^{3-} is frequently the limiting nutrient for algal growth in lakes (Schindler, 1977), PO_4^{3-} loading from septic systems is of concern when surface-water bodies occur nearby. Precipitation of vivianite [$Fe\ (PO_4)_2 \cdot 8H_2O$] in lake bottom sediments has been noted at an important control on PO_4^{3-} levels in lakes (Nriagu and Dell, 1974). Chemical equilibrium modeling using the computer code PHREEQE (Parkhurst et al., 1980), indicates that at the enriched Fe levels given in Tables 2-4, (4.2-39 mg/l), vivianite saturation would be reached at PO_4^{3-}-P levels of 0.03-0.7 mg/l. These are relatively low concentrations representing only 1-10 percent of effluent values. Indeed, in some environments, such a barrier's ability to limit PO_4^{3-} might be considered more important than its ability to attenuate NO_3^-.

Figure 5 suggests that excess carbon leaching will occur at least during initial startup and will be a function of barrier residence time. After one year of operation, DOC levels in pore water exiting the barriers varies from 86 mg/l at the Killarney site (Table 2) where residency is longest (40 days), to <5 mg/l at the Long Point site (Figure 6) where residency is shortest (10 days). DOC levels of tens of mg/l, however, can be commonly found in pore water of other organic-rich environments, for example, in the O and A horizons of upland forest soils in the northeastern United States (20-40 mg/l, Cronan and Aiken, 1985; McDowell and Likens, 1988; Qualls et al., 1991) and in some aquifers which contain labile organic material such as peat (Aravena and Wassenaar, 1993). It is also likely that much of the excess DOC will be relatively immobile as a result of sorption onto Fe oxyhydroxide and clay minerals present in downgradient sediments (Jardine et al., 1989; Dunnivant et al., 1992). DOC that is absorbed downgradient may, in fact, act as a reservoir of carbon available for denitrification as carbon from the barrier becomes depleted. Thus, generation of excess DOC may not be a serious concern in some cases.

It is expected that the two design options proposed for the denitrification barrier, horizontal layer or vertical wall, would be used in different situations. A barrier layer can be economically installed with new septic systems at the time of the tile bed construction, while the wall configuration may be more useful for retrofitting existing septic systems where NO_3^- contamination has already occurred. In areas where the water table is relatively deep, difficulty of installation may render the wall configuration impractical; however, the layer configuration can be used at any water-table depth provided the barrier porous media is appropriately sized to remain tension-saturated when positioned above the water table. The wall configuration can be complemented by other technologies such as the "funnel-and-gates" concept of Starr and Cherry (1994). In this remediation scheme, a plume is "funnelled," using impermeable sheet pile, to a localized point of treatment (gate) where the reactive material is contained. This configuration allows for more convenient manipulation and replenishment of the reactive media as conditions warrant.

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