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Factors influencing nitrate depletion in a rural stream

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Abstract

A mass balance procedure was used to analyze rates of nitrate depletion in three adjacent reaches of West Duffin Creek, Ontario, Canada. Daily nitrate losses in individual reaches were highly variable (0.5–24 kg N) during low and moderate stream flows in May–October, 1982–1985. Nitrate removal efficiency (nitrate loss as a % of nitrate input) showed a rapid exponential decline with increased nitrate inputs to each reach. Nitrate losses and nitrate removal efficiency also had a significant negative correlation with stream discharge. The association of large nitrate loads with high stream discharge reduced the nitrate removal capacity of the stream because of shorter residence times and a higher ratio of water volume to stream bed area. Water temperature exhibited a significant positive correlation with nitrate loss which may reflect increased denitrification at higher temperatures.

Variations in nitrate losses and nitrate removal efficiency between the three reaches were highly influenced by differences in water residence time. Standarized nitrate losses with respect to water residence time revealed a longitudinal decline in nitrate depletion between the reaches which was associated with a downstream decrease in stream nitrate concentration and in the organic carbon content of fine textured sediments from pool habitats.

Introduction

Although recent studies of nutrient transport and transformations have contributed to a greater appreciation of streams as dynamic ecosystems, the factors that influence nitrate depletion in streams draining forest and agricultural landscapes are not well understood. Stream budget studies have shown considerable seasonal and annual nitrate losses during transport in some agricultural streams (Kaushik & Robinson, 1976; Hill, 1979, 1983; Hoare, 1979; Cooper & Cooke, 1984). Annual nitrate losses also occurred in a small forest stream particularly following disturbance by clearcutting (Swank & Caskey, 1982). In contrast, other investigations revealed an absence of significant nitrate depletion in several forest and agricultural streams (Johnston *et al.*, 1976; Triska *et al.*, 1984; Richey *et al.*, 1985).

Nitrate losses in some New Zealand streams were attributed to macrophyte uptake (Howard-Williams *et al.*, 1982; Cooper & Cooke, 1984). However, most studies have identified denitrification in anaerobic stream sediments as the major mechanism of nitrate depletion (Chatarpaul & Robinson, 1979; Swank & Caskey, 1982; Hill & Sanmugadas, 1985). Laboratory experiments with stream and lake sediments have demonstrated that microbial denitrification is affected by temperature and the nitrate concentration of the overlying water (Terry & Nelson, 1975; Toms *et al.*, 1975; Sain *et al.*, 1977; Van Kessel, 1977). The availability of organic matter as a metabolizable energy source often exerts a dominant influence on denitrification rates in soils (Knowles, 1982). In laboratory experiments it has been shown that organic matter in stream sediments influences the denitrifaction rate (Kaushik *et al.*, 1981). Hill & Sanmugadas (1985) found that denitrification rates in sediments from three Ontario streams were more highly correlated with water-soluble organic carbon than with total organic carbon. Rates of denitrification may also be enhanced by the presence of oligochaete worms in sediments (Kaushik *et al.*, 1981). These laboratory studies provide an indication of the various factors that have a potential to regulate denitrification but there is little data available regarding the relative importance of these factors in the stream system. Nitrate depletion in streams can also be influenced by hydrological characteristics. Radiotracers and nutrient enrichment experiments indicate that flow regime and water residence time can affect the retention capacity of streams with respect to phosphorus (Meyer, 1979; Mulholland *et al.*, 1985).

This paper examines the influence of environmental factors on nitrate loss during transport in Duffin Creek, Ontario. Previous research had shown that denitrification was a major mechanism of nitrate depletion in this stream (Hill, 1981; Hill & Sanmugadas, 1985). Consequently, I evaluated factors such as water temperature, stream nitrate concentration and sediment organic carbon content that affect rates of denitrification, as well as stream flow regime and water residence time. Interactions between these factors may be particularly important in determining the capacity of streams to deplete nitrate.

Study area

Duffin Creek is a shallow hard-water stream which flows into Lake Ontario 30 km east of Toronto. The three stream reaches selected for mass balance analysis are located on the West Duffin Creek and extend downstream for approximately 15 km from the junction of Reesor Creek and West Duffin Creek to station 5 (Fig. 1). The West Branch of Duffin Creek drains a mixed farming landscape with corn forming one of the main crops. Stouffville, a town with a population of about 5000 people is served by a tertiary sewage treatment system which discharges effluent into Reesor Creek about 9 km upstream from site 1.

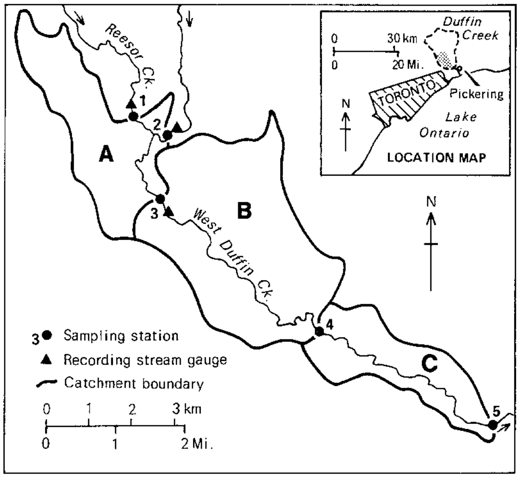

Fig. 1. West Duffin Creek showing location of sampling stations, stream gauges and stream reaches for which budgets were calculated.

The stream in the study is composed of alternating riffles and pools and the average channel width increases from 4 to 5 m at sites 1 and 2 to 10 m at site 5. Water depth varies from 2–20 cm in the riffles and from 20–100 cm in pools during low summer flows. Substrate materials range from gravel and cobbles on riffles to calcareous sand and silt in pool habitats. Portions of the channel north of site 3 are shaded by riparian forest, elsewhere the channel is unshaded and flows through abandoned pastures and scrubland. Aquatic macrophytes are almost entirely absent from the stream and the main component of the flora consists of *Cladophora* mats on the gravel riffles.

Methods

A mass balance procedure was used to construct a nitrogen budget for three reaches of the stream. A chloride mass balance was also examined because this element is a mobile anion similar to nitrate but not subject to removal by physical or biological processes during stream transport. During low to moderate stream flow conditions the stream nutrient load as it enters and leaves each reach constitutes the major input and output terms in the budget. Additional inputs may be represented by small tributary streams and subsurface flow entering the study reaches. A mass balance approach cannot detect recycling or nutrient 'spiralling' (Newbold *et al.*, 1981) of nitrogen within a stream reach. Denitrification in deeper anaerobic sediments and nitrification in the oxygenated water column and aerobic surface sediments may occur concurrently in streams (Chatarpaul *et al.*, 1980). Consequently the mass balance provides a minimum estimate of nitrate depletion in the stream.

Research was conducted during the May to October months of 1982–85. Previous research indicated that nitrate removal occurs mainly during these months in Duffin Creek (Hill, 1981). Moreover, the inaccuracy of discharge measurements on ice-covered streams and element inputs from unsampled ephemeral tributaries create considerable errors in mass balance calculations during the winter months (Hill, 1983). The analysis of a mass balance was restricted to days with low to moderate stream flows. Occasional storm events during the May–October period were not sampled because it was impossible to measure contributions from small ephemeral tributaries and saturated areas adjacent to the stream channel.

Stream discharge and element concentrations were measured at sites 1 to 5. Stream flow recording gauges provided discharges for sites 1 to 3 and current meter measurements coupled with cross-sectional areas were used to calculate discharge at sites 4 and 5. The latter procedure was also used to estimate flows entering the study reaches from several small tributaries during early May and late October. During the summer months these tributaries ceased flowing. There are no major inputs of ground water in the downstream reaches of West Duffin Creek (Sibul *et al.*, 1977). However, small gravel terraces contribute a minor amount of subsurface flow to the study reaches. The chemistry of this subsurface flow was estimated by collecting water samples from 10–12 seeps on several occasions during the study.

Water samples were analyzed for nitrate using an automated cadmium reduction method (APHA, 1976). Tests were also made separately for nitrite in the water samples. Since the concentration of nitrite was very low the nitrate + nitrate values will be referred to as nitrate. Water samples were analyzed for ammonium using an automated indophenol blue method (Technicon, 1975). Total soluble nitrogen was measured by a potassium persulfate oxidation method (Solorzano & Sharp, 1980). Soluble organic nitrogen was obtained by subtraction of the ammonium and nitrate + nitrite from the total soluble nitrogen.

The data used to calculate mass balance were collected at irregular time intervals between early May and late October. The analysis was based on totals of 40 (reach A), 47 (reach C) and 63 (reach B) observations dates. Dye tracing was used to measure the time of travel of water through the study reaches at low and moderate discharges. These estimates of travel time were then used to sample the chemistry of the same parcel of water as it passed through each reach. Water temperature was measured at sites 1 to 5 at times of water sampling. These temperatures were averaged for input and output stations to provide a mean temperature for each reach.

The organic carbon content of stream bed sediments was assessed in June and July 1985. Fine textured sediments located within 1 m of the stream bank were sampled in pool segments at 15 locations spaced at intervals of approximately 1 km between sites 1 and 5. At each location a plexiglass tube was used to remove a 0–5 cm depth sediment sample from the stream bed at 5 randomly selected sites along a 20 m section of the channel. Organic carbon in the sediment was estimated by the Walkley-Black method (Allison, 1965). Water-soluble organic carbon in the sediments was determined by analyzing extracts obtained by shaking 30 g of air-dried sediment with 60 ml of water for 30 min and centrifuging and filtering the resulting suspension through a 0.45 μm membrane filter. Organic carbon in the filtrate was estimated following the procedure of Waring & Gilliam (1983).

Results

Nitrogen and chloride budgets

Low concentrations of ammonium (<0.05 mg N L^{-1}) and trace levels of nitrite occurred at the stream sampling sites. Concentrations of nitrate usually ranged between 1.4 and 2.3 mg N L^{-1} at site 1, 0.8–1.2 mg N L^{-1} at sites 2 and 3 and 0.5–1.0 mg N L^{-1} at sites 4 and 5. Soluble organic nitrogen varied between 0.1 and 0.3 mg N L^{-1} at sites 2 to 5 and 0.4–0.6 mg N L^{-1} at site 1. High chloride concentrations ranging between 30–50 mg L^{-1} were recorded at site 1, whereas levels were 6–7 mg L^{-1} at site 2 and 16–20 mg L^{-1} at sites 3 to 5. Ground water entering the study reaches from seeps contained less than 0.1 mg NO_3-N L^{-1} and low levels of chloride (<6 mg L^{-1}).

The water budget for the three study reaches showed that average daily inputs and outputs of water were approximately in balance (Table 1). These water budgets suggest that the contribution of subsurface flow to the reaches is very small. Moreover, as the seeps contain very low concentrations of nitrate, the subsurface component has been ignored in the calculation of daily elements budgets. The nitrate budget for the three reaches showed that inputs exceeded outputs on all observation days during 1982–85. Mean daily deficits ranged from 5 kg NO_3-N in reach C to 14.1 kg NO_3-N in reach B. Daily deficits were particularly large during low stream flows in July and August. Under these conditions, nitrate depletion in reach B was often 16–24 kg N d^{-1}, a range of loss which represented 50–75% of the daily nitrate input. Analysis of soluble organic nitrogen transport indicated no consistent pattern of gains or losses and the average daily difference between inputs and outputs was about 3–6% for the three reaches (Table 1). The absence of a significant difference between chloride inputs and outputs is in contrast to the considerable loss of nitrates and suggests that the procedures and measurements used in the mass balance analysis are reasonable.

Factors influencing nitrate depletion in individual reaches.

During the study period stream discharge ranged from about 300 L s^{-1} in mid-summer to 1300–1400 L s^{-1} in early May and late October. Daily nitrate inputs to each reach varied from 20–30 kg N to a maximum of 160 kg N, whereas

Table 1. Mean (±SD) daily input and output of water, nitrogen and chloride for three reaches of West Duffin Creek during May–October 1982–85.

	Input	Output	Net gain or loss	Gain or loss (% of input)
Reach A (N = 40)				
Flow (L s^{-1})	541 ± 199	549 ± 215	+8 ± 20	1
NO_3-N (kg d^{-1})	59.9 ± 25.6	50.4 ± 21.7	−9.5 ± 3.1**	16
Soluble organic N (kg d^{-1})	11.1 ± 3.9	10.6 ± 3.5	−0.5 ± 2.7	5
Chloride (kg d^{-1})	781 ± 192	766 ± 239	−15 ± 85	2
Reach B (N = 63)				
Flow (L s^{-1})	547 ± 239	568 ± 262	+21 ± 31	4
NO_3-N (kg d^{-1})	53.4 ± 26.3	39.2 ± 29.1	−14.1 ± 4.3**	26
Soluble organic N (kg d^{-1})	10.7 ± 4.7	11.4 ± 4.3	+0.7 ± 4.1	6
Chloride (kg d^{-1})	774 ± 251	803 ± 293	+29 ± 93	4
Reach C (N = 47)				
Flow (L s^{-1})	571 ± 250	593 ± 266	+22 ± 24	4
NO_3-N (kg d^{-1})	40.9 ± 22.5	36.0 ± 23.5	−5.0 ± 1.6**	12
Soluble organic N (kg d^{-1})	11.2 ± 4.1	11.5 ± 4.0	+0.3 ± 3.4	3
Chloride (kg d^{-1})	813 ± 290	829 ± 314	+16 ± 64	2

**Mean daily gain or loss significantly different from zero at the 1% level using a pair comparison t test.

water temperature ranged between 6° and 27 °C.

Daily nitrate losses in each reach had a significant but weak negative correlation with daily nitrate inputs (Table 2). Consequently, the nitrate removal efficiency of the stream measured in terms of daily loss as a percentage of daily inputs exhibited a rapid exponential decline. For example in reach A removal efficiency decreased from about 30% at inputs of 30–40 kg NO_3-N d^{-1} to less than 5% at inputs of >100 kg NO_3-N d^{-1} and reach B showed a decline from 70% to less than 15% over a similar range of inputs.

With respect to the environmental factors evaluated in this study, daily nitrate losses showed a relatively high positive correlation with water temperature (Table 2, Fig. 2). Stream discharge had a significant negative correlation with daily nitrate loss and the nitrate removal efficiency of the individual reaches also exhibited an exponential decline as stream discharge increased (Fig. 3). No significant relationship was observed between nitrate loss and nitrate concentration of inputs to reaches A and B, wheras a weak negative correlation between these variables occurred in reach C.

A number of the factors evaluated showed a considerable degree of interdependence (Table 3). Water temperature was negatively correlated with daily nitrate input and stream discharge, particularly in reaches A and B. These interrelationships reflect the seasonal pattern of discharge, water temperature and nitrate flux in West Duffin Creek. Larger nitrate inputs occurred during periods of higher discharge and cooler temperature in May and October rather than in summer. Daily nitrate inputs were also highly correlated with increases in stream discharge but had a weaker positive correlation with nitrate concentration (Table 3). Consequently larger nitrate inputs were associated with a decrease in water residence time.

Table 2. Correlation coefficients between daily nitrate loss and stream variables for each stream reach.

Stream variable	Daily NO_3-N loss		
	Reach A	Reach B	Reach C
NO_3-N input (X_1)	−0.47**	−0.60**	−0.65**
Water temperature (X_2)	0.70**	0.73**	0.70**
NO_3-N concentration (X_3)	−0.02	−0.11	−0.33*
Discharge (X_4)	−0.54**	−0.64**	−0.68**

*Significant at the 0.05 level; **significant at the 0.01 level.

Contrasts between reaches in nitrate depletion

The three reaches examined had considerable differences in mean daily nitrate losses (Table 1). Nitrate removal efficiencies also showed major contrasts during periods of similar discharge. For example, removal efficiencies were 30–40% in reaches A and C and 70–80% in reach B at daily discharges of 300–400 L s^{-1}.

These differences must be interpreted with caution because the three stream reaches vary in length and stream bed area and therefore have different water residence times. When mean daily nitrate loss was calculated on a stream bed area basis the highest loss occurred in reach A (407 mg N $m^{-2}d^{-1}$), and depletion decreased in a downstream direction to 233 and 99 mg N $m^{-2}d^{-1}$ in reaches B and C respectively. However a comparison of the reaches

Table 3. Correlation matrix for stream variables for each stream reach (stream variables $X_1 - X_4$ are listed in Table 2).

	X_1	X_2	X_3	X_4
Reach A				
X_1	1	−0.73**	0.48**	0.88**
X_2		1	−0.27	−0.70**
X_3			1	0.06
X_4				1
Reach B				
X_1	1	−0.68**	0.45**	0.87**
X_2		1	−0.31*	−0.66**
X_3			1	−0.01
X_4				1
Reach C				
X_1	1	−0.62**	0.71**	0.89**
X_2		1	−0.46**	−0.48**
X_3			1	−0.36**
X_4				1

*Significant at the 0.05 level; **significant at the 0.01 level.

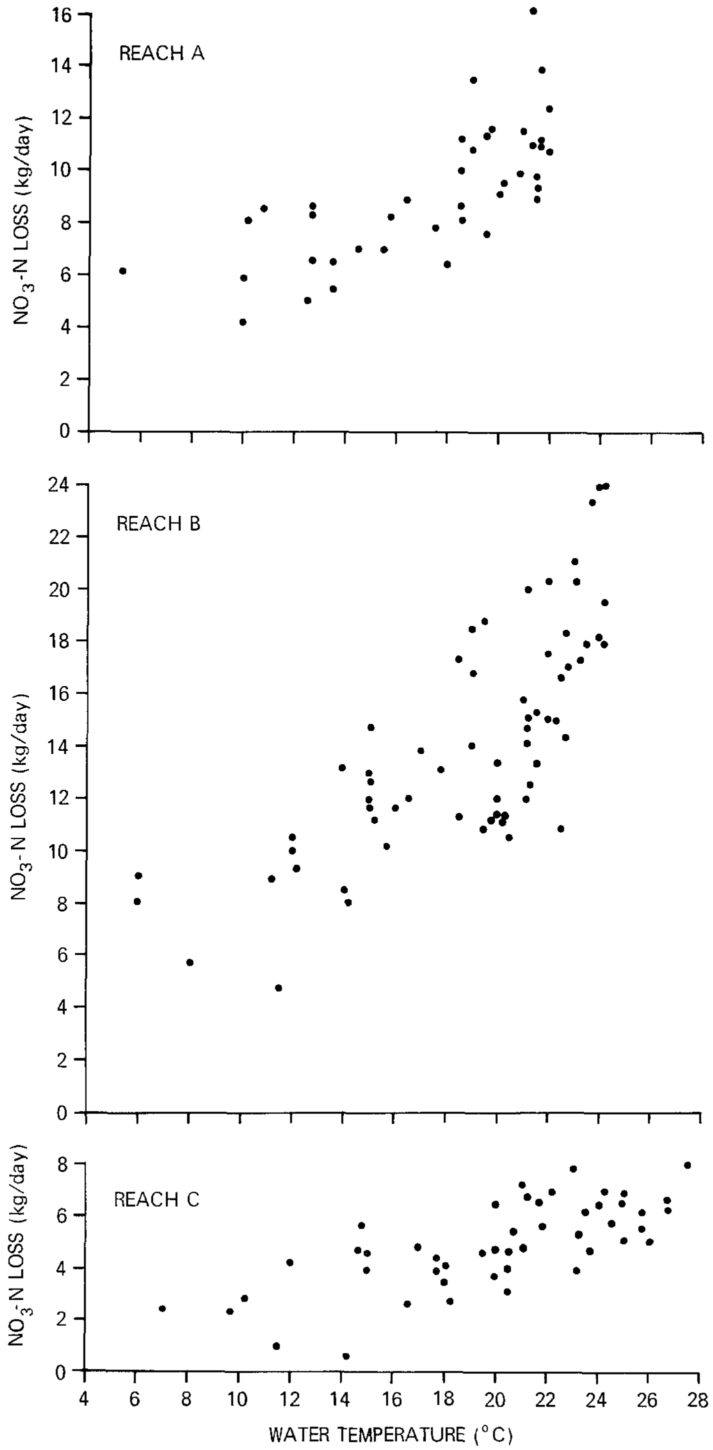

Fig. 2. Relationships between nitrate losses and water temperature for the three stream reaches.

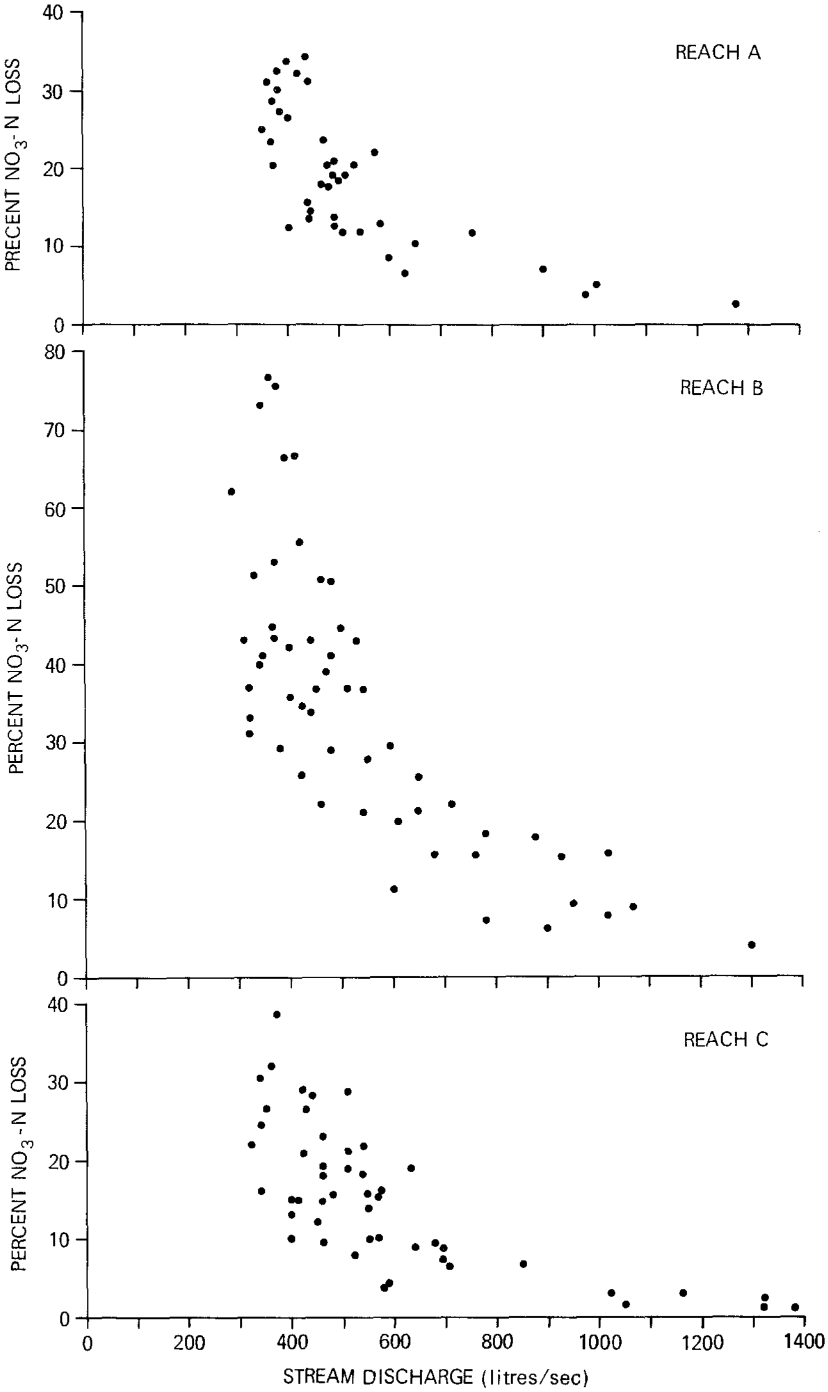

Fig. 3. Relationships between nitrate removal efficiency and stream discharge for the three stream reaches.

on an areal basis may not accurately reflect differences in water residense time because this factor can be affected by variations in channel gradient and water depth.

The dye-tracing data provided an estimate of water residence time in each reach over a range of discharges. For example, at discharges of about 400 L s^{-1} water travel times were approximately 2.5, 7 and 4 hours for reaches A, B and C respectively. Nitrate depletion rates were standardized with respect to flow-through time in order to account for differences between reaches in water residence time. This was done by calculating daily nitrate losses for individual observation dates based on a one hour flow-through time. Standardised daily nitrate depletion rates were usually less than 2 kg N in reach C, 1.5–3 kg N in reach B and 3–6 kg N in reach A (Fig. 4). These data indicate that the higher nitrate losses observed in reach B in comparison to reaches A and C resulted from the longer residence time of water (Table 1).

Differences in standardised nitrate losses between the reaches suggest that other factors besides water residence time may influence longitudianal variations in nitrate depletion. Downstream variations in water temperature occurred particularly in July and August when temperatures in reach C were often 4–5 °C higher than in reach A (Fig. 2). Despite this difference the standardised nitrate losses were much greater in reach A than in C. Stream nitrate concentration showed a significant but weak positive correlation with standardised nitrate losses for each reach ($r = 0.55$, $P < 0.01$) (Fig. 4). However, depletion rates often differed considerably between reaches despite similar nitrate concentrations suggesting that other factors are also important in determining nitrate loss.

Total and water-soluble organic carbon in fine textured pool sediments adjacent to the stream banks showed considerable variability nevertheless, mean values declined in a downstream direction (Fig. 5). In particular total and water-soluble organic carbon were about 3 times greater in reach A and the initial 3 km of reach B than in reach C. Although these data do not represent average stream bed carbon levels, they provide an estimate of carbon for stream habitats which are probably sites of maximum denitrification activity.

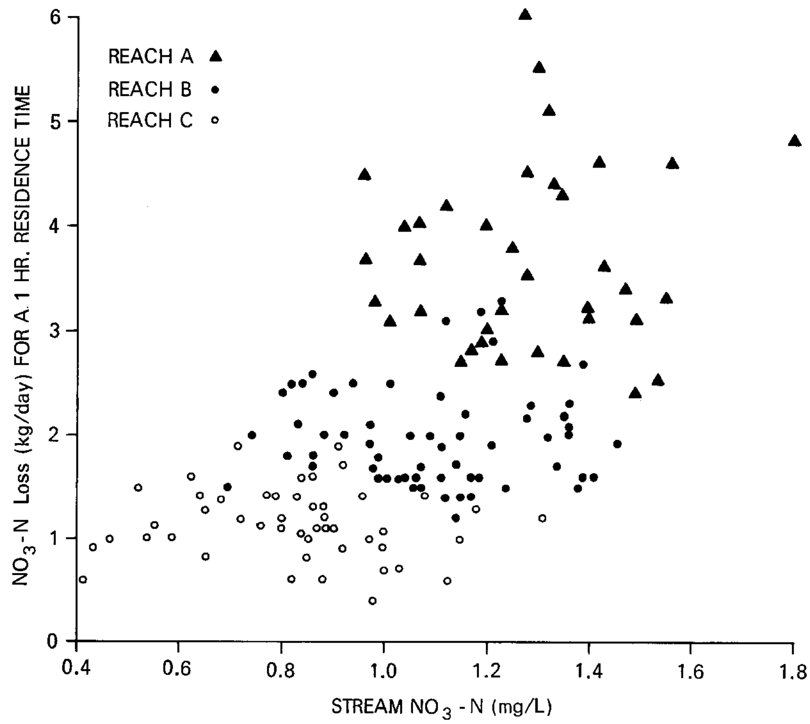

Fig. 4. Relationships between standardised nitrate loss and the initial nitrate concentration of water inputs to each reach.

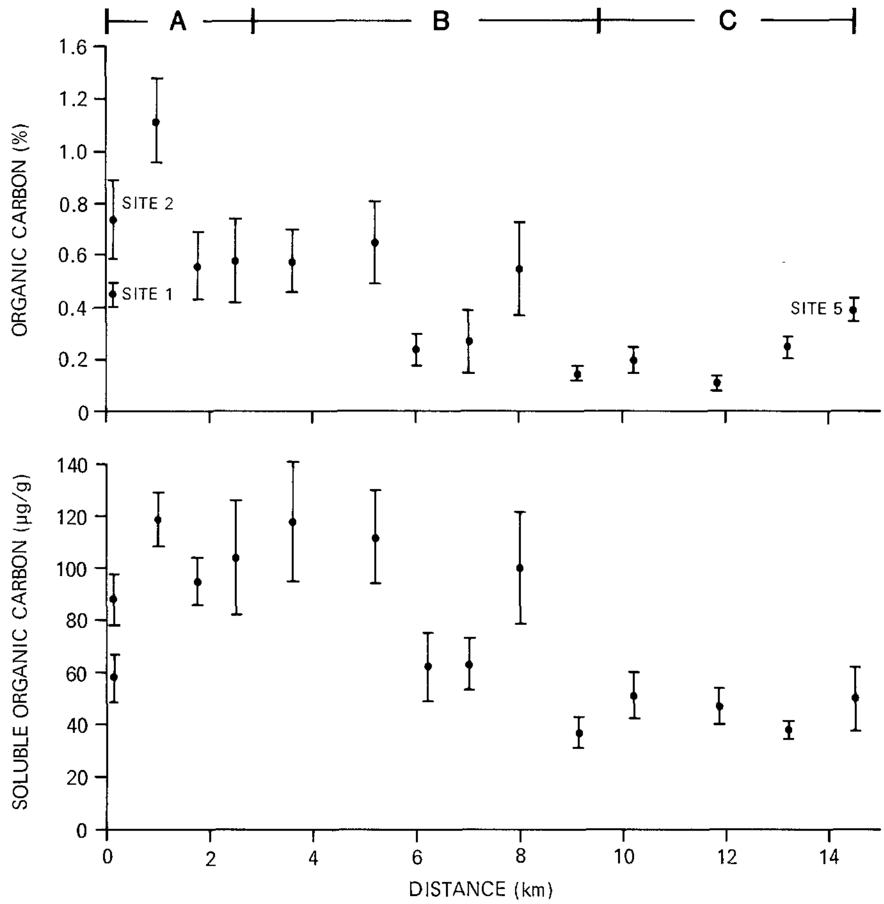

Fig. 5. Variations in total and water soluble organic carbon in 0–5 cm depth sediments from pool habitats in West Duffin Creek. (Data are means ± 1 SE for 5 measurements at each site.)

Discussion

Relationships between nitrate load, nitrate losses and environmental factors suggest several possible reasons for variations in nitrate depletion rates in individual reaches. The positive correlation between water temperature and daily nitrate loss is probably influenced partly by the effect of temperature on rates of denitrification. The temperature coefficient (Q_{10}) is generally in the range of 1.5 to 3.0 for denitrification in soils (Knowles, 1982). Laboratory incubations of stream sediments have shown that nitrate loss at temperatures of 10–11 °C was 50–60% of the loss at 20–22 °C (Sain *et al.*, 1977; Hill, 1983).

Water temperature and stream discharge were negatively correlated, thus, the effect of temperature on nitrate depletion cannot be clearly separated from the influence of discharge. Variations in stream discharge may affect nitrate losses and the efficiency of nitrate depletion for several reasons. Nitrate entering stream reaches during periods of low discharge has a longer residence time and is more likely to undergo biological transformations. The dye-trace data suggest that the flow-through time in each reach increased by >100% as discharge declined from about 1300 to 300 L s^{-1}. A second factor which may reduce removal is the greater water depth at high discharges which decreases the probability of contact between a nitrogen molecule in the water column and the sediment surface (Huang & Wozniak, 1981). An increase in the velocity gradient at the sediment-water interface may create greater sediment disturbance at higher discharges. This disturbance could increase the depth of the aerobic surface layer in the stream sediment inhibiting denitrification and possibly enhancing rates of nitrification. An increase in water velocity could also remove particulate organic matter from the surface of the sediment. Such particles may provide micro sites for denitrification. Although denitrification rates may be reduced during periods of higher stream discharge it should be emphasized that a decline in stream nitrate removal efficiency is not dependent on decreased nitrate losses. An increase in nitrate inputs associated with higher discharges would produce a decline in removal efficiency even though the denitrifying capacity of the stream reach remained constant or increased by an amount which represented a lower proportion of the nitrate input.

Lower removal efficiencies for phosphorus and carbon at higher stream discharges have also been reported for stream reaches (Meyer & Likens, 1979; Minshall *et al.*, 1983; Mulholland *et al.*, 1985). However, the small number of observations collected in these studies precluded a detailed assessment of the relationship between discharge and ecosystem efficiency. Meyer & Likens (1979) suggested that the relationship between water residence time and the proportion of nutrient inputs retained in a stream reach may exhibit a threshold residence time above which there will be no increase in the fraction of nutrient input which enters biogeochemical pathways. The pattern of nitrate removal efficiency in relation to stream discharge in West Duffin Creek indicates that this threshold is not attained, even at very low summer discharges. It appears that rates of nitrate depletion in relation to stream residence time are too low to attain a threshold in this stream.

Denitrification may show a positive association with nitrate concentration in streams (Hill, 1981; Duff *et al.*, 1984; Triska *et al.*, 1984). Stream nitrate concentrations in reaches A and B were usually >0.6 mg N L^{-1} and varied over a relatively narrow range of concentration which probably accounts for the lack of significant correlation with nitrate depletion (Table 2). The significant but weak negative correlation between nitrate concentration and daily nitrate loss in reach C probably reflects the tendency for high nitrate concentrations to be associated with cooler water temperatures and larger stream discharges, factors which reduce nitrate depletion.

A comparison of the three reaches indicates that contrasts in daily nitrate removal were highly influenced by differences in water residence time. Nevertheless, when nitrate depletion rates were standardised with respect to water residence time nitrate losses showed a progressive downstream decline in the three reaches. The association of this longitudinal trend in standardised nitrate losses with a decrease in stream nitrate concentration and organic carbon in pool sediments suggest that these factors may be responsible for differences in nitrate depletion. However, it should be noted that unmeasured variables such as sediment depth and the distribution of benthic fauna could also influence downstream trends in standardised nitrate losses.

One clear conclusion from this study is that nitrate removal efficiency in West Duffin Creek declined rapidly as nitrate inputs increased. This pattern of ecosystem efficiency was influenced mainly by the association of large nitrate fluxes with increased stream discharge and shorter water residence times. In many streams discharge fluctuates over 4 or 5 order of magnitude whereas nitrate concentration remains more constant, although frequently displaying seasonal variations (Likens *et al.*, 1977; Webb & Walling, 1985). Thus transport of nitrate in these streams is strongly influenced by stream discharge.

The proportion of seasonal and annual nitrate load transported by high stream discharges is probably a key factor in influencing the magnitude of nitrate depletion in relation to stream nitrogen budgets. Streams exhibit considerable differences in the relative proportion of annual runoff which is contributed by 'quickflow' (storm flow) and by base flows. Walling (1971) found that the storm flow contribution to annual runoff varied from 15 to 50% in 5 catchments in S. W. England. A high proportion of stormflow occurs in catchments with thin soils and impermeable parent materials, whereas baseflow is favoured by geology and soils which promote rapid infiltration and by the presence of extensive aquifers which store large amounts of ground water (Walling, 1971). Although few stream nitrate budgets have been reported, it is noteworthy that approximately 68% of the annual nitrate input was removed in a small New Zealand stream in which storm flows accounted for only 10% of the total annual runoff

(Hoare, 1979). In contrast, about 60% of the annual nitrate flux in Duffin Creek occurs during high flows in the February-April snowmelt period and nitrate removal only represents 5–6% on the annual nitrogen export (Hill, 1979). A greater understanding of the significance of nitrate depletion requires field studies of nitrogen mass balances in a wide variety of rivers and associated efforts to relate patterns of depletion to environmental factors.

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