Exploring functional similarity in the export of nitrate-N from forested catchments: A mechanistic modeling approach

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Abstract. Functional similarity of catchments implies that we are able to identify the combination of processes that creates a similar response of a specific characteristic of a catchment. We applied the concept of functional similarity to the export of NO$_3^-$-N from catchments situated within the Turkey Lakes Watershed, a temperate forest in central Ontario, Canada. Despite the homogeneous nature of the forest, these catchments exhibit substantial variability in the concentrations of NO$_3^-$-N in discharge waters, over both time and space. We hypothesized that functional similarity in the export of NO$_3^-$-N can be expressed as a function of topographic complexity as topography regulates both the formation and flushing of NO$_3^-$-N within the catchment. We tested this hypothesis by exploring whether topographically based similarity indices of the formation and flushing of NO$_3^-$-N capture the observed export of NO$_3^-$-N over a set of topographically diverse catchments. For catchments with no elevated base concentrations of NO$_3^-$-N the similarity indices explained up to 58% of the variance in the export of NO$_3^-$-N. For catchments with elevated base concentrations of NO$_3^-$-N, prediction of the export of NO$_3^-$-N may have been complicated by the fact that hydrology was governed by a two-component till, with an ablation till overlying a basal till. While the similarity indices captured peak NO$_3^-$-N concentrations exported from shallow flow paths emanating from the ablation till, they did not capture base NO$_3^-$-N concentrations exported from deep flow paths emanating from the basal till, emphasizing the importance of including shallow and deep flow paths in future similarity indices. The strength of the similarity indices is their potential ability to enable us to discriminate catchments that have visually similar surface characteristics but show distinct NO$_3^-$-N export responses and, conversely, to group catchments that have visually dissimilar surface characteristics but are functionally similar. Furthermore, the similarity indices provide a potentially powerful method to scale and generalize NO$_3^-$-N export responses to other regions.

1. Introduction

Catchments are considered to be functionally similar if different combinations of the process controls result in the same response; the change in one process control is compensated for by a change in another process control. The uniqueness of the concept of functional similarity is that first, it provides a method that will enable us to state, a priori, the interactions of processes that will create the same response and, second, it provides a mechanistic basis for interpretation of the observed differences in the response (E. M. O'Loughlin, unpublished manuscript, 1991).

In this paper, we apply the concept of functional similarity to the export of NO$_3^-$-N from catchments situated within the Turkey Lakes Watershed (TLW), a temperate forest in central Ontario, Canada. Despite a narrow range in the loading of N to the individual catchments, there is a wide range in the loss of N from the forest to surface waters [Creed, 1998]. These catchments exhibit substantial variation in the concentrations of NO$_3^-$-N in the discharge waters, over both time and space [Nicolson, 1988; Creed, 1998]. In our application of the concept of functional similarity to the export of NO$_3^-$-N we attempt to understand the underlying mechanisms that determine why NO$_3^-$-N export from these catchments varies so dramatically.

In a recent paper [Creed et al., 1996] we derived similarity indices for representing the dominant processes controlling the export of NO$_3^-$-N. These indices were tested on a single catchment within the Turkey Lakes Watershed. The indices captured processes regulating both the formation of NO$_3^-$-N within the catchment surface and the flushing of NO$_3^-$-N from the catchment to the receiving waters. For the single catchment the indices were able to capture the NO$_3^-$-N peaks in discharge waters under distinct catchment functional states. The indices captured NO$_3^-$-N peaks that occurred during spring melt or autumn storms, when a rising water table rose into previously unsaturated parts of a NO$_3^-$-N-rich soil profile. The indices also captured N peaks that occurred during summer droughts, when a rising water table rose into previously saturated parts of a NO$_3^-$-N-poor soil profile following a period of enhanced rates of nitrification [Creed et al., 1996]. Although the catchment functional states under these two scenarios were distinct, the NO$_3^-$-N exports were similar.

The similarity indices were based on the doctrine that topography regulates NO$_3^-$-N formation and NO$_3^-$-N flushing processes. Topography can be an important regulator of nu-
trient cycling and routing dynamics as it influences (1) the formation of nutrient sink areas (areas that retain N) and source areas (areas that release N to adjacent waters) within the catchment, (2) the hydrologic flushing of these nutrient source and sink areas, and (3) the export of nutrients accompanying this formation and flushing [Murdoch and Stoddard, 1992; Stoddard, 1994; Hornberger et al., 1994; Boyer et al., 1995, 1996; Creed et al., 1996].

In this paper, we hypothesize that NO$_3^-$-N export is a function of the topographic complexity of the catchment as topography regulates both NO$_3^-$-N formation processes and NO$_3^-$-N flushing processes within the catchment. To test this hypothesis, we explore whether the similarity indices capture the observed variation in the export of NO$_3^-$-N over a set of catchments representing the range in topographic complexity within the watershed.

2. Test Area

The test area is the Turkey Lakes Watershed (TLW), an old-growth sugar maple forest located in the Algoma Highlands of central Ontario, Canada. Situated on the Canadian Shield, this 10.5 km$^2$ watershed contains a headwater chain of lakes fed by small catchments that ultimately drain into Batchawana Bay on the eastern shoreline of Lake Superior (Figure 1).

The TLW was selected as the test area because potential sources of variation in the export of NO$_3^-$-N were few. The selection criteria mandated that the test area should (1) be within a single climatic region; (2) be within a single forest region; (3) contain a forest dominated by a single species; (4) contain a forest within a stable stage of development; (5) contain a substantial range in the hypothesized topographic controls on the export of NO$_3^-$-N; and (6) have sufficient data to test the hypotheses. The TLW is located within the Superior Climatic Region (criterion 1) and the Algoma Section of the Great Lakes-St. Lawrence Forest Region (criterion 2). The forest is dominated by sugar maple (Acer saccharum Marsh.), with small stands of yellow birch (Betula alleghaniensis Britton) and other minor species (criterion 3). The forest is an unmanaged (120–180 years) mature forest that with the exception of a light harvest of yellow birch over 35 years ago, remains essentially undisturbed (criterion 4). Given the homogeneous character of the catchments in terms of criteria 1–4, we were able to focus on the topographic effects on the export of NO$_3^-$-N. The TLW topography is complex. Controlled by bedrock, the undulating topography results in a small, sharply defined subwatersheds for each lake and provides substantial heterogeneity of topographic features both within and among the catchments (criterion 5).

Since the autumn of 1980 the TLW has served as a continuous monitoring site for the study of catchment hydrology and biogeochemistry (see references in a special issue on TLW published in Canadian Journal of Fisheries and Aquatic Sciences, 45, supplement 1, 1988). A catchment monitoring network was strategically established to encompass the range in the complexity of the catchment topography, with some catchments characterized by relatively gentle, concave slopes and other catchments characterized by relatively steep, convex slopes. Previously unpublished data collected from this monitoring network during the 10-year period from 1981 to 1990 provided the basis for this investigation on the topographic effects on the export of NO$_3^-$-N (criterion 6).

3. Methods

In our conceptual model (Figure 2) the export of NO$_3^-$-N is a function of the hydrologic flushing of NO$_3^-$-N [Hornberger et al., 1994; Creed et al., 1996]. On the basis of this conceptual model, when the water table is low, N accumulates within the soil profile, resulting in small export of NO$_3^-$-N to adjacent waters. As the water table rises, it "flushes" the soil profile, and NO$_3^-$-N within the soil profile is available for export. As the water table reaches the soil surface, the water table flushes the organically enriched surface layers of the soil profile, resulting in potentially large export of NO$_3^-$-N to adjacent waters, unless denitrification processes become significant. To test our conceptual model, we adopted a "hybrid" modeling approach in which we simulated indices that represent the hypothesized controls on the export of NO$_3^-$-N and related these indices to the observed export of NO$_3^-$-N.

3.1. RHESSys Simulation System

The Regional HydroEcological Simulation System (RHESSys) (Figure 3) is a distributed model designed to compute the distribution of biogeochemical stores and the fluxes within catchments. The distributed model captures the variability in biogeochemical processes by subdividing the catchment into hillslopes. Within each hillslope a probability distribution function (pdf) of catenary positions is calculated. Catenary positions differ in their properties because of downslope transport processes that include (1) differential drainage conditions, (2) differential transport and deposition of suspended materials, and (3) differential leaching, translocation and redeposition of soluble materials [Hall and Olson, 1991]. Catchment biogeochemical fluxes are determined through simulation at two scales: the hillslope scale, where simulation over the pdf of catenary positions considers the variability in biogeochemical fluxes related to the nonlinear impacts of wetter and drier parts of the hillslope, and the catchment scale, where simulation over the population of hillslopes considers the variability in biogeochemical fluxes related to differences in the average slope, aspect, or elevation of the hillslopes within the catchment. For a more detailed description of RHESSys the reader is referred to Running and Gower [1988], Running and Goward [1991], Running and Hunt [1993], and Band et al. [1991, 1993].

3.1.1. RHESSys data inputs. A combination of distributed and nondistributed data inputs is required to drive the RHESSys simulation system (Figure 3). For each catchment the drainage outlet was located within a Global Positioning System–derived Universal Transverse Mercator coordinate system. The drainage area was extracted and partitioned into hillslopes, ridge lines, and stream lines according to the methods of Band [1989], Lammers and Band [1990], and Mackay and Band [1994]. Once delimited, hillslopes were parameterized for forest, topographic, and soil attributes.

Distributed canopy leaf area index (LAI) data were derived from LANDSAT TM data [Nemani et al., 1993] that were calibrated using a combination of DECAGON Sunfleck Centimeter–derived and allometrically derived LAI data. Topographic attributes, including both primary (slope, aspect, elevation, contributing area) and secondary (catenary index) attributes, were derived from digital terrain analysis [Band, 1986, 1989]. Soil attributes, including effective mineral soil depth (or rooting depth), saturated surface hydraulic conductivity $K_s$, and the rate of change of saturated surface hydraulic
conductivity with soil depth \( m \), were estimated from field- and literature-based data. Distributions of \( K_s \) and \( m \) in catchments that range from 5 to 70 ha were difficult to estimate, particularly as field data were collected from sparse sites. Although a realistic range of these parameters was estimated from the field and the literature, the actual values within this range were determined during model calibration in which the simulated discharge was matched to the observed discharge. Using the same set of parameters for \( K_s \) and \( m \), we produced acceptable model simulations of discharge hydrographs (simulated versus observed estimates of annual discharge were within \( \pm 15\% \), with no apparent bias among the years) for each catchment for the period from 1981 to 1990 [Creed et al., 1996; I. F. Creed, unpublished data, 1996].
3.1.2. N source index: The N source index represents the availability of NO$_3^-$-N in the soil profile. A simple N source index is based on the assumption that N cycling activity is stoichiometrically related to C cycling activity. This assumption is reasonable for the TLW soils which are characterized as having an almost 1:1 transformation of organic N to inorganic N [Foster, 1985, 1989]. The N source index is computed as the ratio of C supply to C demand. The C supply is an exponential function of the maximum rate of gross organic C decomposition modified by a moisture factor (MF) and a temperature factor (TF) [after Parton et al., 1987]. The C demand is a function of net carbon fixation (equation (1)).

$$\text{Source} = \frac{\text{Supply}}{\text{Demand}} = \frac{C_{\text{potential soil pool}}(1 - \exp \{MF \times TF\})}{C_{\text{photosynthesis}} - C_{\text{respiration}}}$$ (1)

Nondistributed forest physiological parameters were derived from field- and literature-based data and are summarized by Creed et al. [1996]. The meteorological variables driving the simulation system, including daily precipitation and daily minimum and maximum temperature data, were collected at an Atmospheric Environment Service (AES) meteorological recording station located at the southeast boundary of the TLW. Missing data were estimated by linear regression on the basis of data from the closest meteorological stations within the area for which complete records were available (Montreal Falls and Sault Sainte Marie, Ontario). The meteorological variables were extrapolated from the recording station to each hillslope using the RHESSys climate model, MTCLIM [Running et al., 1987], enabling us to capture variations due to slope, aspect, and elevation but not variations due to topographic shadows.

3.1.2. RHESSys data outputs. RHESSys simulation outputs provide the basis for computation of similarity indices that represent the hypothesized controls on the export of NO$_3^-$-N (Figure 3): (1) a N source index, which considers the availability of NO$_3^-$-N to be flushed from the soil surface, and (2) a N flush index, which considers the flushing of NO$_3^-$-N caused by the interactions of the water table with NO$_3^-$-N within the soil profile. The RHESSys-derived N indices provide a means of exploring N dynamics both at the subcatchment scale, in terms of potential sinks and sources of NO$_3^-$-N within the catchment, and at the catchment scale, in terms of the potential export of NO$_3^-$-N from the catchment. As the focus of this paper is to compare results among different catchments, we derived N indices from catchment-average dynamics. A short description of the derivation of the N source and flush indices used by Creed et al. [1996] is given below, and the reader is referred to that reference for additional detail.

Figure 2. A conceptual model of the hypothesized controls on the export of NO$_3^-$-N. The flushing export of NO$_3^-$-N is a function of two groups of interactive processes: (a) processes that control the net accumulation of N in the soil, and (b) processes that control the net transport of N from the soil. The flushing export of NO$_3^-$-N is presented in three diagnostic stages. At stage 1 the water table is low and water-soluble forms of N accumulate within the soil profile, resulting in small concentrations of NO$_3^-$-N in discharge waters. At stage 2, as the water table starts to rise, water-soluble forms of N in the soil profile are flushed, resulting in moderate concentrations of NO$_3^-$-N in discharge waters. At stage 3, as the water table continues to rise, water-soluble forms of NO$_3^-$-N in the organically enriched surface of the soil profile are flushed, resulting in large concentrations of NO$_3^-$-N in discharge waters.

Figure 3. A schematic diagram of the Regional HydroEcological Simulation System (RHESSys). The engine of RHESSys is the simulation system. Data inputs to the simulation system include distributed forest, terrain, and soil data and nondistributed climate data and forest physiological parameters. Data outputs from the simulation system include water and carbon budgets, from which are derived the N source and N flush indices.
To examine the variability of the N source index, both within and among catchments, a relative N source index is computed that normalizes the source index to the watershed's temporal and spatial maximum N source index (equation (2)).

\[
\text{Relative Source Index} = \frac{\text{Source}}{\text{Source}_{\text{max}}} \quad (2)
\]

### 3.1.2.2. N flush index:

The N flush index represents the interactions of the water table with NO\textsubscript{3}-N within the soil profile. The water table dynamics are based on the soil water saturation deficit. Soil water saturation occurs when all the pores in the soil are filled with water. Although a deficit is typically represented on a negative scale, in this paper, we present its absolute value, on a positive scale. The hillslope-average saturation deficit \(S_h\) is computed on the basis of the following water balance equation (equation (3)):

\[
S_h = S_p - R_n + Q \quad (3)
\]

where

- \(S_h\) hillslope-average soil saturation deficit, m;
- \(S_p\) previous day hillslope-average soil saturation deficit, m;
- \(R_n\) net recharge to the saturated zone, a function of the throughfall or melt that exceeds the interception capacity of the litter and the unsaturated soil water dynamics as computed by BIOME-BGC, m;
- \(Q\) hillslope discharge to the stream, m.

In catchments with significant topographic relief the distribution of soil water can be highly heterogeneous, leading to nonlinear effects on the biogeochemical cycling rates within the catchment [Band, 1993; Band et al., 1993]. Topographic attributes have been used to estimate the distribution of soil water within a hillslope [Beven and Kirkby, 1979; O'Loughlin, 1981]. In TOPMODEL [Beven and Kirby, 1979], the distribution of water within the hillslope is dependent on catenary position. Catenary positions with larger contributing drainage areas and smaller surface slopes (i.e., near streams) will have small saturation deficits, while catenary positions with smaller contributing drainage areas and larger surface slopes (i.e., near ridges) will have large saturation deficits. For each point \(i\) within the hillslope a catenary index is computed (equation (4)) on the basis of the assumption of a steady state subsurface throughflow system in which recharge from the unsaturated to the saturated zone in the contributing area above a unit length of contour is equal to the saturated throughflow across the contour length [Beven and Kirby, 1979; Beven, 1986].

\[
\text{CI} = \ln \left( \frac{a T_c}{T_i \tan \beta} \right) \quad (4)
\]

where

- \(\text{CI}\) catenary index, m\(^2\);
- \(a\) contributing area, m\(^2\);
- \(T_c\) local soil transmissivity, m/d;
- \(T_i\) hillslope-average soil transmissivity, m/d;
- \(\beta\) local gradient, degrees.

The catenary index is used to compute the saturation deficit at point \(i\), \(S_i\) (equation (5)):

\[
S_i = S_h + m(\lambda - \text{CI}) \quad (5)
\]

where

- \(S_i\) local soil saturation deficit, m;
- \(S_h\) hillslope-average soil saturation deficit, m;

\(m\) a parameter proportional to the rate of change of saturated hydraulic conductivity with depth; \(
\lambda\) hillslope-average catenary index (\(\int \ln (a T_c/T_i \tan \beta) \, da\)).

Biogeochemical stores and fluxes, including evaporation, transpiration, photosynthesis, and respiration are computed for each catenary position within a catchment. At the end of each day the saturation deficit for each catenary position is area-weighted to update the saturation deficit for each hillslope \(S_3\) and, in turn, this saturation deficit is area-weighted to update the saturation deficit for the catchment \(S\), which forms the basis of the N flush index.

The N flush index is computed as \(S/S_{30}\), the ratio of the current saturation deficit to the previous 30-day average saturation deficit. While \(S\) considers only the current day saturation deficit, \(S/S_{30}\) incorporates \(S\), the rising and receding oscillations of \(S\), and the frequency of the oscillations of \(S\). The N flush index provides three possible scenarios: (1) If \(S/S_{30} < 1\), the current day water table is higher than before, and saturated throughflow is rising into previously unsaturated parts of the soil profile (i.e., catchment is flushing). (2) If \(S/S_{30} > 1\), the current day water table is lower than before, and saturated throughflow is receding (i.e., catchment is draining), and (3) if \(S/S_{30} = 1\), there is no change in the water table relative to the previous 30-day period. The 30-day average saturation deficit is an appropriate length of time between consecutive flushings for the accumulation of substantial N in the soil profile for the Turkey Lakes Watershed [Creed et al., 1996]; however, the optimal length of time will vary over the year. In addition, the optimal length of time will vary for different regions. Note that since \(S\) represents the catchment's average saturation deficit, intersection of a rising water table with the NO\textsubscript{3}-N within the soil profile can occur either when \(S\) is small (water table is high, and large flushing areas within the catchment are present) or when \(S\) is large but one tail of the distribution of soil saturation deficits is small (water table is low, but small flushing areas within the catchment are present).

### 3.2. Chemistry of Discharge Waters

For each catchment, total daily water discharge (mm/d) was derived from a continuously measured stream-gauge station equipped with a 90° or 120° V notch weir. Missing data (usually winter small flows) were estimated by linear regression with an adjacent catchment for which there was a complete record. Stream water samples were collected every 2 weeks during the winter, daily during spring snowmelt, and every week or every 2 weeks during the summer and autumn. Samples were collected at the same sampling point, in the center of the stream, during each visit. To remove particulate matter, each sample was filtered through a Whatman No. 41 filter that had been rinsed with distilled water. To determine the concentration of NO\textsubscript{3}-N, a 100-mL subsample was filtered through a 0.45-μm Millipore membrane filter that had been washed with distilled water and stored without preservative at 2°C in glass containers that had been washed with acid water (0.1 N H\textsubscript{2}SO\textsubscript{4}) and rinsed with distilled water. The subsample was analyzed for NO\textsubscript{3}-N within 48 hours of collection by a cadmium reduction procedure using a Technicon AutoAnalyzer II-C+. To determine the concentration of calcium (Ca), a 100-mL subsample was stored without preservative at 2°C in polyethylene containers that had been washed with acid water (10% HNO\textsubscript{3}) and rinsed with distilled water. The subsample was analyzed for Ca within 7 days of collection by atomic absorption on a Varian 1275 spectrophotometer [Nicolson, 1988].
4. Results and Discussion

4.1. Nitrate-N Export Behavior

Nitrate-N export could be classified according to one of four types of responses (Figure 4) [Creed, 1988]: NO$_3$-N peaks restricted to spring melt with small NO$_3$-N base concentrations (Figure 4a), NO$_3$-N peaks occurring throughout the year with small NO$_3$-N base concentrations (Figure 4b), NO$_3$-N peaks occurring throughout the year with elevated NO$_3$-N base concentrations (Figure 4c), and NO$_3$-N peaks restricted to the spring melt with elevated NO$_3$-N base concentrations (Figure 4d). We hypothesized that the observed variation in the export of NO$_3$-N among these catchments was due to factors intrinsic to each catchment (e.g., topographic complexity) rather than factors extrinsic to the catchments. We attempted to capture the observed variation in the export of NO$_3$-N with similarity indices that capture both the intrinsic and extrinsic processes involved in the export of NO$_3$-N from catchments.

4.2. Similarity in N Dynamics

By using our hybrid modeling approach, we generated response surfaces that represent the observed export of NO$_3$-N as a function of the simulated hypothesized controls on the export of NO$_3$-N (Figure 5). An analysis of these surfaces provides insights to the importance of the hypothesized controls. If NO$_3$-N export is restricted to catchment conditions reflecting a combination of NO$_3$-N formation and NO$_3$-N flushing activities, then we were successful at capturing the dominant controls on NO$_3$-N export. However, if NO$_3$-N export is not restricted to these catchment conditions, then other processes are important for NO$_3$-N export.

The assemblage of catchments shows similar responses in the export of peak concentrations of NO$_3$-N (Figure 5, light
Similarity in the "single" peak in the N response surface that occurs in response to high N flushing and N formation activities.
The form of the response surfaces suggests that the concentration of \( NO_3^-\)N in discharge waters may be an exponential function of the N source and flush indices (equation (6)):

\[
N = a \exp(-\beta FI) \exp(+\gamma SI)
\]

where

- \( N \) \( NO_3^-\)N concentration, mg/L;
- \( SI \) Source Index, dimensionless;
- \( FI \) Flush Index, dimensionless;
- \( a, \beta, \gamma \) empirical coefficients.

For c50, a catchment with minimal base concentrations of \( NO_3^-\)N (where base concentrations reflect \( NO_3^-\)N export during a nonflushing state), a \( NO_3^-\)N export model based on an exponential function of the N indices provides results that are significant, but not with a large degree of explanation (Figure 7, \( p < 0.001 \), multiple \( r^2 = 0.32 \)). We attribute the small predictive power largely to limitations of the N source index that are described below.

### 4.2.1. Immobilizing versus mobilizing properties

A seasonal analysis of the residuals of the \( NO_3^-\)N export model (Figure 8) indicates that the export of \( NO_3^-\)N during the autumn flushing time period was overestimated. For the N source index (equations (1) and (2)) a weakness of the computation of the supply term is that in assuming a stoichiometric relationship between the generation of carbon (CO2) and N, we did not consider the role of microbial dynamics in the generation of N. In particular, we did not discriminate between the immobilization (i.e., organic N) versus mobilization (i.e., \( NO_3^-\)N) fates of the mineralized nitrogen. Although the assumption of a stoichiometric relationship for the generation of CO2 and N (i.e., a 1:1 transformation of organic N to inorganic \( NO_3^-\)N) applies over most of the season [Foster, 1989], the assumption does not apply with canopy turnover in the autumn season.

With canopy turnover, there is a major input of relatively large C/N organic matter to the forest floor. During the early...
stages of decomposition, heterotrophic microbes flourish and, although they produce CO₂ in the decomposition process, they consume NH₄⁺-N to grow. Upon microbial mortality a portion of the consumed NH₄⁺-N is converted into forms that compose the humus complex, and a portion is again mineralized into NH₃-N forms available for microbial uptake. Ammonium-N is immobilized and therefore nitrification, the oxidation of NH₃-N to NO₃⁻-N, is inhibited. During this stage of decomposition the dramatic reduction in NH₃-N results in a concomitant reduction in NO₃⁻-N in the soil waters [Foster, 1989; Foster et al., 1992; Yin et al., 1993]. As decomposition continues, the C/N ratio of the organic matter declines as C is lost and N is conserved, slowly accumulating mineral N. Eventually, the reduced microbial demand for mineral N results in a significant accumulation of NO₃⁻-N concentrations in the soil solution of the winter months [Foster, 1985; Yin et al., 1993], until the next flush, often occurring during the spring melt period [Creed et al., 1996].

During the canopy turnover period our N source index is large, reflecting a large supply term (rapid rates of carbon digestion) and a small sink term (low rates of carbon fixation) (equation (1)). Although autumn storm-induced flushing activity will increase the concentrations of NO₃⁻-N in the discharge waters, the predicted increase is dampened as a significant portion of the N pool is in an immobilized, rather than mobilized, form that is not available for flushing.

If we do not include data collected during the autumn season, the predictive power of the NO₃⁻-N export model increases to 58% (multiple \( r^2 = 0.58 \), significance \( p < 0.001 \), Figure 9). Given the simplicity of this model of NO₃⁻-N export, we are encouraged by these results. Inclusion of the C/N factor to discriminate between the immobilization versus mobilization fate of mineral nitrogen to enable inclusion of autumn data will be considered in future research.

### 4.2.2. Instantaneous versus inheritance properties

Another limitation of the N source index is that it does not consider the impact of the memory of the catchment in terms of N dynamics. The memory of a catchment is an important control on both the concentrations and the fluxes of NO₃⁻-N in discharge waters [Burt et al., 1988]. On an annual basis a system may shift between “flush-limited” and “source-limited” states with respect to the peak concentrations of NO₃⁻-N during the dominant flushing period. A system is characterized as flush-limited if a small frequency and magnitude of flushing results in small peak concentrations of NO₃⁻-N as the pool of N is stored within the catchment. In contrast, a system is characterized as source-limited if a large frequency and magnitude of flushing results in small peak concentrations of NO₃⁻-N as the pool of N within the catchment is exhausted. The potential impacts of memory on the establishment of flush- versus source-limited states and the impacts of these states on the export of NO₃⁻-N from c50 are explored below.

A chronosequence of the export of peak concentrations of NO₃⁻-N for c50 is presented in Figure 10. A water year was defined as the period from June to the following May, enabling us to follow the fate of the annual cohort of N from its dominant periods of formation during the summer and winter [Foster, 1985, 1989; Yin et al., 1993] through to its dominant period of flushing during the following spring [Creed et al., 1996]. The chronosequence represents a transition from dry to wet summer conditions over 7 water years. For the 1983 and 1984 water years, characterized by dry summers, the peak concentrations of NO₃⁻-N were relatively small. For the subsequent water years, characterized by wet summers, the peak concentrations of NO₃⁻-N were initially relatively large (1985, 1986, and 1987) and then small (1988 and 1989) (Figure 10).

We hypothesize that while the export of NO₃⁻-N may be similar along different points of the chronosequence, the functional state of the catchment may be quite different (Figure 11). Along the chronosequence of NO₃⁻-N export, during the 1983 and 1984 water years, the catchment was flush-limited, resulting in relatively small peaks in NO₃⁻-N export. During the relatively dry summers of 1983 and 1984, N accumulated, creating an enhanced N pool within the catchment. The forms of N that accumulated were largely immobile forms as the dry
Figure 11. A conceptual model of the effect of climate on peak concentrations of exported NO$_3^-$-N. The size of the arrow represents the relative peaks in the export of NO$_3^-$-N during a sequence of relatively dry to wet water years. For the relatively dry water years, 1983 to 1984, the catchment is flush-limited, resulting in relatively small peak concentrations in exported NO$_3^-$-N. For the transition to relatively wet water years, 1985 to 1987, the catchment is neither flush- nor source-limited, resulting in relatively large peak concentrations in exported NO$_3^-$-N. For the wet water years, 1988 to 1989, the catchment is source-limited, resulting in relative small peak concentrations in exported NO$_3^-$-N (refer to the text for further details).

conditions inhibited the microbial conversion of N to mobile forms [Foster, 1989; Foster et al., 1992]. Hill and Shackleton [1989] suggest that summer droughts reduce N mineralization and nitrification rates and this reduction in NO$_3^-$-N formation rates may continue into the following winter. On the basis of a 50-year record of climate the summers of 1983 and 1984 were exceptionally dry. During these droughts the immobile N pool was likely augmented not only by the accumulation of NH$_4^+$-N due to the inhibition of nitrification processes [Foster et al., 1992] but also by the accumulation of organic N due to the desiccation of the finer root systems of the forest. For the subsequent 1985, 1986, and 1987 water years the catchment was neither flush- nor source-limited, resulting in relatively large peaks in NO$_3^-$-N export. The enhanced N pool started to mobilize, and a significant pool of NO$_3^-$-N was available for export. The relatively large peaks in NO$_3^-$-N export were sustained for several years following the drought, perhaps reflecting a lag in the mobilization of the N pools created during the drought summers. Eventually, the lag effect disappeared as the enhanced N pool was exhausted. The influence of climatic variability and, in particular, climatic extremes has been reported to have a significant, long-lasting effect on the concentrations of NO$_3^-$-N in discharge waters [Reynolds and Edwards, 1995]. Finally, for the 1988 and 1989 water years, the catchment was source-limited, resulting in relative small peaks in NO$_3^-$-N export as the N pool returned to predrought concentrations. At the start and end of the chronosequence the catchment functional states were distinct, but the NO$_3^-$-N export was similar. While this discussion focuses on climate-induced lags on the transformation of soil N pools from immobile to mobile forms, clearly, the forest itself will also respond to climate, with its own lag [Braswell et al., 1997], potentially altering the supply to and/or the demand from the soil N pools. This hypothesis needs to be tested in the field.

The instantaneous dynamics of the source- versus flush-limited states of the catchment are considered in the similarity indices, confirming the importance of the interactions of the source and flush controls on the export of NO$_3^-$-N. However, to strengthen the effectiveness of the indices, we need to incorporate the history of the dynamics of the mobile and immobile N pools in the soils.

4.2.3. Configuration properties. According to our NO$_3^-$-N export model (equation (6)), NO$_3^-$-N that is flushed from contributing source areas within the catchment is efficiently exported to the discharge waters. In reality, significant barriers may exist to the efficient export of NO$_3^-$-N, including subsequent infiltration as the flushed NO$_3^-$-N passes through unsaturated areas en route to the catchment outlet and/or denitrification as the flushed NO$_3^-$-N passes through saturated areas contiguous to the catchment outlet. We hypothesize that the contributing source areas of NO$_3^-$-N to discharge waters will vary dynamically depending on their degree of organization within the catchment [Creed and Band, 1997]. Organization is defined as the connection of source areas to the catchment outlet. Organized source areas are hydrologically connected to the catchment outlet, and hydrologic pulses transmit N efficiently to the outlet. In contrast, disorganized source areas are isolated and hydrologically disconnected from the outlet. In these isolated source areas, under most hydrologic conditions, NO$_3^-$-N is retained, requiring large hydrologic pulses to transmit the NO$_3^-$-N to the outlet. The potential importance of the organization of source areas is demon-
strated in c50. For this catchment the relationship between the saturation deficit and the residuals of the model (model based on Figure 9) is presented in Figure 12. When the saturation deficit is 0, the residuals range from -2 to +2. When the saturation deficit increases, however, the residuals range from -4 to +2. The negative bias in the residuals suggests that as the saturation deficit increased, isolated source areas that were not contiguous and were not connected to the outlet were formed. In this catchment the isolated areas may have created barriers to the efficient export of NO$_3$-N. This hypothesized influence of organized versus disorganized NO$_3$-N source areas needs to be tested using a combination of model- [Creed and Band, 1997] and field-based experiments.

### 4.3. Dissimilarity in N Dynamics

While the assemblage of catchments show similar responses in the export of peak concentrations of NO$_3$-N (light areas), they show dissimilary responses in the export of base concentrations of NO$_3$-N (dark areas) (Figure 5). The dissimilarity among the catchment responses can be seen in the gradient of the base concentrations of NO$_3$-N, which range from 0 to 0.5 mg/L (Figure 5).

One hypothesis is that this gradient in the base concentrations of NO$_3$-N reflects a differential contribution of NO$_3$-N from the deep hydrologic flow paths among the catchments. The TLW bedrock is overlain by a surficial till that consists of two components: a more permeable ablation till overlying a less permeable basal till. At low to moderate elevations the ablation till overlies the basal till at a depth of about 0.5 m, while at high elevations the ablation till overlies bedrock as there is no basal till [Craig and Johnston, 1988; Nicolson, 1988]. This surficial till structure results in the formation of shallow and deep hydrologic flow paths. During a hydrologic event, water enters the surficial till and percolates vertically and then laterally along the interface with the bedrock or, more commonly, along the interface with the basal till [Bottomley et al., 1984, 1986]. A water table or perched water table forms, rising and flushing NO$_3$-N from the soil profile [Foster, 1985]. As shown in other temperate forests in North America, a portion of the water that enters the surficial till may be conducted through macropores [Peters et al., 1995] to the basal till, translocating N from shallow to deep flow paths [Burns et al., 1993], where it is released over time, months to years later. During hydrologic events the dominant source of exported NO$_3$-N may be from deep flow paths emanating from the ablation till. In between hydrologic events the dominant source of exported NO$_3$-N may be from deep flow paths emanating from the basal till (Figure 13). According to our hypothesis, there are differences in the extent of basal till among the catchments. The two extremes are catchments with no basal till (i.e., catchments with no contribution from deep flow paths; for example, c50) and catchments completely covered with basal till (i.e., catchments with the greatest contribution from deep flow paths; for example, c39) (Figure 5).

To test this hypothesis, we used a natural chemical tracer to reflect the flow paths of NO$_3$-N through the tills. Catchment discharge waters will reflect the relative amounts of NO$_3$-N that emanate from each of the tills if the composition of the ablation and basal tills is distinct. In the surficial till the concentration of calcite (CaCO$_3$) is smaller in the ablation till than in the basal till [Kusmirski and Cowell, 1983; Johnston and Craig, 1987]. Two explanations for the smaller concentrations of calcite in the ablation till are (1) differential weathering of the ablation and basal tills (calcite was weathered faster from the ablation till because of the generation of preferential flow paths at the ablation-basal interface of the tills) [Kusmirski and Cowell, 1983] and (2) different provenance of the ablation and basal tills [Karrow and Geddes, 1987]. To support the use of Ca as a chemical tracer, we examined the relationship between Ca (mg/L) and the depth to the water table (represented as a saturation deficit S) (Figure 14). The relationship depicted suggests that as the depth to the water table increases (i.e., as the catchment system shifts from shallow to deep flow paths), the proportion of water in contact with Ca-rich till increases, and therefore the concentration of Ca in the discharge waters increases. Although the Ca chemistry provides an indication of the source of discharge waters, the Ca chemistry of the discharge waters will change both over time (during large flows the water table will be located within the Ca-poor till, while during small flows the water table will be located within the Ca-rich till) [Rice and Bricker, 1995] and over space (catchments will have varying amounts of the Ca-rich till).

The ratio of NO$_3$-N/Ca provides an indication of the relative source of NO$_3$-N in discharge waters. For a given concentration of NO$_3$-N, discharge waters dominated by shallow flow paths will be characterized by small Ca concentrations (short residence time in the more permeable, Ca-poor till) and a large ratio of NO$_3$-N/Ca. In contrast, large Ca concentrations (long residence time in less permeable, Ca-rich till) and a small ratio of NO$_3$-N/Ca will characterize discharge waters dominated by deep flow paths (Figure 13).

For all of the catchments the slopes of the NO$_3$-N versus NO$_3$-N/Ca relationship based on seasonal and annual groupings of the data are presented in Table 1. As expected, the regression coefficients of the NO$_3$-N versus NO$_3$-N/Ca relationships were typically smaller for small flow periods (sum-
LARGE flow periods

SMALL flow periods

No deep hydrologic flow paths, No NO\textsubscript{3}-N export

Shallow flow paths through the ablation till are the dominant source of NO\textsubscript{3}-N during large flow periods

Deep flow paths through the basal till are the dominant source of NO\textsubscript{3}-N during small flow periods

☐ Ablation Till (Small Ca, Large NO\textsubscript{3}-N/Ca)

☐ Basal Till (Large Ca, Small NO\textsubscript{3}-N/Ca)

Figure 13. A conceptual model of the role of hydrologic flow paths on the export of NO\textsubscript{3}-N. Catchments vary in the amount of basal till present. For all catchments, shallow flow paths through the ablation till are the dominant source of NO\textsubscript{3}-N during large flow periods. For those catchments with no basal till, there is no export of NO\textsubscript{3}-N during small flow periods. For those catchments with basal till, deep hydrologic flow paths through the basal till are the dominant source of NO\textsubscript{3}-N during small flow periods.

mer, reflecting a greater relative contribution from deep flow paths) than for large flow periods (spring, reflecting a greater relative contribution from shallow flow paths) [Rice and Bricker, 1995]. However, the range in the regression coefficients of the NO\textsubscript{3}-N versus NO\textsubscript{3}-N/Ca relationships was comparatively narrow within a catchment and broad among the catchments (Table 1). For a representative subset of the catchments (including the four catchments depicted in Figures 4 and 5) the NO\textsubscript{3}-N versus NO\textsubscript{3}-N/Ca relationships based on all available data are presented in Figure 15. Assuming our interpretation of the NO\textsubscript{3}-N versus NO\textsubscript{3}-N/Ca relationship is correct, NO\textsubscript{3}-N from catchments c50 and c46 emanates primarily from shallow flow paths, while NO\textsubscript{3}-N from catchments 39 and c31 emanates from both deep and shallow flow paths (Figure 15; notice that the order of catchments in Figure 15 follows the same order of catchments in Figures 4 and 5). The differential dominance of the hydrologic flow paths among the catchments is important, as streams draining catchments dominated by shallow hydrologic flow paths are expected to be more responsive to N additions [Ross et al., 1994].

Catchment c37 is the single catchment that does not follow the conceptual model presented in Figure 13. Catchment c37, together with c50, represent "Type A" NO\textsubscript{3}-N export responses, characterized as catchments with a single peak in the concentrations of exported NO\textsubscript{3}-N during spring melt and no base concentrations of exported NO\textsubscript{3}-N (Figure 4). Although the NO\textsubscript{3}-N export responses are similar for these catchments, the regression coefficient for the NO\textsubscript{3}-N/Ca versus NO\textsubscript{3}-N relationship is highest for c50 and among the lowest for c37, indicating different flow paths taken by the exported NO\textsubscript{3}-N. What can explain this difference? Previous field observations at the TLW provide clues to explaining the apparent anomaly found in c37. In their calculations of lake water budgets, Jeffries et al. [1988] reported a water budget deficit for Wischart Lake and suggested that outgoing seepage through the bottom sediments of this lake to Little Turkey Lake could have contrib-
Figure 14. The relationship of Ca (mg/L) as a function of the depth to the water table (represented as the saturation deficit S) for the four catchments depicted in Figures 4 and 5. The concentration of Ca in the discharge waters increases as the depth to the water table increases, indicating that the concentration of Ca in the discharge waters increases as the source of the discharge waters shifts from shallow to deep flow paths.

In his calculation of stream water budgets, Nicolson [1988] reported that the unusually high flows from c37 may have originated from the subsurface seepage from Wischart Lake to Little Turkey Lake, as part of this seepage could have been intercepted and discharged as surface flow from c37. This seepage would have been characterized by high Ca concentrations, as weathering of calcite and dolomite [Craig and Johnston, 1988] controls the chemistry of groundwaters in the TLW. Therefore, while fast flow paths may be the dominant source of NO\textsubscript{3}-N export in c37, the use of the NO\textsubscript{3}-N/Ca ratio may be obscured by the addition of NO\textsubscript{3}-N-poor, Ca-rich subsurface seepage waters. These are speculations, and until further field observations are collected, the flow paths by which NO\textsubscript{3}-N reaches the discharge waters of c37 will remain unresolved.

On the basis of our analysis of the hydrologic flow paths, a more appropriate model of NO\textsubscript{3}-N export would have the following structure:

\[ N = SFP + DFP = \alpha \exp(-\beta S) + \chi + DFP \]  

Table 1. Annually and Seasonally Derived Regression Coefficients of the NO\textsubscript{3}-N Versus NO\textsubscript{3}-N/Ca Relations

<table>
<thead>
<tr>
<th>Catchment</th>
<th>Annual</th>
<th>Summer</th>
<th>Autumn</th>
<th>Winter</th>
<th>Spring</th>
<th>( r^2 ) Annual</th>
<th>Summer</th>
<th>Autumn</th>
<th>Winter</th>
<th>Spring</th>
</tr>
</thead>
<tbody>
<tr>
<td>c31</td>
<td>0.17</td>
<td>0.13</td>
<td>0.15</td>
<td>0.15</td>
<td>0.19</td>
<td>0.82</td>
<td>0.74</td>
<td>0.78</td>
<td>0.87</td>
<td>0.74</td>
</tr>
<tr>
<td>c32</td>
<td>0.15</td>
<td>0.09</td>
<td>0.12</td>
<td>0.10</td>
<td>0.16</td>
<td>0.78</td>
<td>0.93</td>
<td>0.39</td>
<td>0.60</td>
<td>0.66</td>
</tr>
<tr>
<td>c33</td>
<td>0.23</td>
<td>0.21</td>
<td>0.23</td>
<td>0.21</td>
<td>0.23</td>
<td>0.93</td>
<td>0.84</td>
<td>0.92</td>
<td>0.82</td>
<td>0.88</td>
</tr>
<tr>
<td>c34</td>
<td>0.22</td>
<td>0.16</td>
<td>0.19</td>
<td>0.18</td>
<td>0.23</td>
<td>0.86</td>
<td>0.88</td>
<td>0.73</td>
<td>0.70</td>
<td>0.84</td>
</tr>
<tr>
<td>c35</td>
<td>0.22</td>
<td>0.19</td>
<td>0.20</td>
<td>0.20</td>
<td>0.23</td>
<td>0.89</td>
<td>0.62</td>
<td>0.59</td>
<td>0.51</td>
<td>0.84</td>
</tr>
<tr>
<td>c37</td>
<td>0.17</td>
<td>0.11</td>
<td>0.13</td>
<td>0.15</td>
<td>0.21</td>
<td>0.77</td>
<td>0.83</td>
<td>0.80</td>
<td>0.78</td>
<td>0.82</td>
</tr>
<tr>
<td>c39</td>
<td>0.12</td>
<td>0.09</td>
<td>0.10</td>
<td>0.10</td>
<td>0.12</td>
<td>0.81</td>
<td>0.69</td>
<td>0.77</td>
<td>0.63</td>
<td>0.55</td>
</tr>
<tr>
<td>c42</td>
<td>0.21</td>
<td>0.19</td>
<td>0.18</td>
<td>0.20</td>
<td>0.22</td>
<td>0.89</td>
<td>0.90</td>
<td>0.75</td>
<td>0.91</td>
<td>0.88</td>
</tr>
<tr>
<td>c44</td>
<td>0.28</td>
<td>0.25</td>
<td>0.29</td>
<td>0.26</td>
<td>0.29</td>
<td>0.93</td>
<td>0.96</td>
<td>0.91</td>
<td>0.86</td>
<td>0.90</td>
</tr>
<tr>
<td>c46</td>
<td>0.31</td>
<td>0.28</td>
<td>0.32</td>
<td>0.29</td>
<td>0.32</td>
<td>0.95</td>
<td>0.88</td>
<td>0.90</td>
<td>0.89</td>
<td>0.91</td>
</tr>
<tr>
<td>c47</td>
<td>0.42</td>
<td>0.38</td>
<td>0.42</td>
<td>0.38</td>
<td>0.43</td>
<td>0.90</td>
<td>0.96</td>
<td>0.93</td>
<td>0.90</td>
<td>0.85</td>
</tr>
<tr>
<td>c49</td>
<td>0.37</td>
<td>0.34</td>
<td>0.37</td>
<td>0.34</td>
<td>0.38</td>
<td>0.94</td>
<td>0.90</td>
<td>0.84</td>
<td>0.97</td>
<td>0.88</td>
</tr>
<tr>
<td>c50</td>
<td>0.46</td>
<td>0.48</td>
<td>0.45</td>
<td>0.43</td>
<td>0.48</td>
<td>0.94</td>
<td>0.89</td>
<td>0.84</td>
<td>0.97</td>
<td>0.90</td>
</tr>
</tbody>
</table>

Annually derived regression coefficients are based on all available data for the period 1982–1990. Seasonally derived regression coefficients are based on all available data for summer (June–August), autumn (September–November), winter (December–February), and spring (March–May) for the period 1982–1990.
SFP NO$_3$-N contributed from shallow flow pathways, mg/L
DFP NO$_3$-N contributed from deep flow pathways, mg/L

With the similarity indices we were able to predict NO$_3$-N export from shallow, fast flow paths, during both the major spring melt and autumn storm hydrologic events and the minor summer hydrologic events. We were not able to predict NO$_3$-N export from deep, slow flow paths. This presents a more significant modeling challenge, as it may require a model that captures the variable contributions of NO$_3$-N from a basal till whose spatial dimensions are difficult to delineate.

5. Conclusions

Nitrate-N export varied among the catchments of the Turkey Lakes Watershed. In some cases the variation was subtle, but in other cases the variation was dramatic. This variation in the export of NO$_3$-N could have been interpreted to mean that these catchments are in different stages of N saturation [Aber et al., 1989; Stoddard, 1994]. According to Stoddard [1994], as a forest catchment progresses from being strongly N-deficient to strongly N-sufficient, the impact on the forest’s N dynamics occurs in four diagnostic stages. At stage 0, NO$_3$-N export is characterized by very small NO$_3$-N concentrations during most of the year with a small episode of large NO$_3$-N concentrations during snowmelt. At stage 1, NO$_3$-N export is still a seasonal phenomenon, but the spring episodes of large NO$_3$-N concentrations in surface waters are larger compared to stage 0. At stage 2 the episodic peaks in NO$_3$-N concentrations are as large as stage 1, but the seasonal pattern is dampened by an increase in the concentrations of NO$_3$-N in base flow. Finally, at stage 3, extremely large NO$_3$-N concentrations and the lack of any significant seasonal pattern in NO$_3$-N concentrations characterize NO$_3$-N export. According to Stoddard’s diagnostic stages of N saturation, depending on which catchment was used to make the diagnosis, the TLW catchments could be diagnosed as being in stage 0 (c37, c50), stage 1 (c42, c44, c46, c47, c49), or stage 2 (c31, c32, c33, c34, c35, c39). However, similarity in the history of N loading to the catchments, the regional climate classification, the forest classification, the forest species, and the forest age all indicate that these catchments are at the same stage of N saturation. This observation emphasizes a potential problem with interpretations based on catchment studies (i.e., paired-catchment analyses) and of scaling studies (i.e., there is a range of responses at the catchment scale that must be integrated to the regional scale).

We applied the concept of functional similarity in the export of NO$_3$-N to determine if an alternative explanation could be found for the variation in NO$_3$-N exports. Functional similarity implies that we are able to identify the processes, their impacts and interactions, that create similar responses in NO$_3$-N exports, and on the basis of these processes we are able to interpret reasons for dissimilar responses in NO$_3$-N exports. In our exploration of the functional similarity in the export of NO$_3$-N, we proposed that NO$_3$-N export is regulated by topography and its regulation of NO$_3$-N formation and flushing processes within the catchment. On the basis of this hypothesis we constructed a NO$_3$-N export model in which observed NO$_3$-N export was a function of simulated indices of NO$_3$-N formation and flushing activities. The NO$_3$-N export model explained a substantial portion of the variation in NO$_3$-N exports among the catchments. The model captured the peak concentrations of NO$_3$-N in discharge waters over all seasons for all catchments, indicating that topographic regulation of the NO$_3$-N formation and flushing areas within the catchment were significant. Improvement to the NO$_3$-N export model will be sought with the inclusion of both temporal dynamics, focusing on the effects of catchment memory on the potential export of NO$_3$-N, and spatial dynamics, focusing on the effects of catchment organization of NO$_3$-N formation and flushing areas on the potential export of NO$_3$-N. Incorporation of these effects will generate a model that can be scaled and generalized with effective explanatory power for peak concentrations of NO$_3$-N. The model did not, however, capture the base concentrations of NO$_3$-N in discharge waters. Model improvement that involves the inclusion of the effects of the configuration and characteristics of "invisible" tills as well as other subsurface features that control catchment processes represents a more challenging problem in the development of models that can be scaled and generalized.

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