INPUT-OUTPUT BUDGETS OF INORGANIC NITROGEN FOR 24 FOREST WATERSHEDS IN THE NORTHEASTERN UNITED STATES: A REVIEW

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Abstract. Input-output budgets for dissolved inorganic nitrogen (DIN) are summarized for 24 small watersheds at 15 locations in the northeastern United States. The study watersheds are completely forested, free of recent physical disturbances, and span a geographical region bounded by West Virginia on the south and west, and Maine on the north and east. Total N budgets are not presented; however, fluxes of inorganic N in precipitation and streamwater dominate inputs and outputs of N at these watersheds. The range in inputs of DIN in wet-only precipitation from nearby National Atmospheric Deposition Program (NADP) sites was 2.7 to 8.1 kg N ha^{-1} yr^{-1} (mean = 6.4 kg N ha^{-1} yr^{-1}; median = 7.0 kg N ha^{-1} yr^{-1}). Outputs of DIN in streamwater ranged from 0.1 to 5.7 kg N ha^{-1} yr^{-1} (mean = 2.0 kg N ha^{-1} yr^{-1}; median = 1.7 kg N ha^{-1} yr^{-1}). Precipitation inputs of DIN exceeded outputs in streamwater at all watersheds, with net retention of DIN ranging from 1.2 to 7.3 kg N ha^{-1} yr^{-1} (mean = 4.4 kg N ha^{-1} yr^{-1}; median = 4.6 kg N ha^{-1} yr^{-1}). Outputs of DIN in streamwater were predominantly NO_3-N (mean = 89%; median = 94%). Wet deposition of DIN was not significantly related to DIN outputs in streamwater for these watersheds. Watershed characteristics such as hydrology, vegetation type, and land-use history affect DIN losses and may mask any relationship between inputs and outputs. Consequently, these factors need to be included in the development of indices and simulation models for predicting 'nitrogen saturation' and other ecological processes.

Keywords: ammonium, input-output relationships, nitrate, nitrogen, nitrogen saturation, watersheds

1. Introduction

In the Temperate Zone of North America, nitrogen (N) is generally considered to

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be a growth-limiting nutrient for terrestrial ecosystems. However, during the past decade it has been proposed that elevated atmospheric N deposition may lead to N saturation, which has been defined as a condition that occurs when the availability of inorganic N is in excess of biotic demand (Aber *et al.*, 1989; Ågren and Bosatta, 1988). If forest ecosystems were to reach this condition, several adverse effects would result, including nutrient imbalances in foliage, increased soil acidification and aluminum mobility, and excess NO_3^- in streams (Aber *et al.*, 1989; Skeffington and Wilson, 1988; Stoddard, 1994). Consequently, elevated N may affect water quality, as well as the productivity and health of forests.

In the northeastern United States, the concern over N saturation has primarily been in response to elevated N deposition associated with acidic deposition. Emissions of N have increased for more than 100 yr, largely as a result of fossil fuel combustion and greater reliance on N fertilizers (Galloway, 1998). Stricter industrial emissions standards have reduced SO_4^{2-} deposition, but N emissions, and hence N deposition, have remained high and relatively constant for the past several decades (Driscoll *et al.*, 2001). Nitrogen amendment studies have shown that forest ecosystems in the northeastern United States have different responses to experimental N inputs (Aber *et al.*, 1995; Adams *et al.*, 1997; Christ *et al.*, 1995; Gilliam *et al.*, 1996; Kahl *et al.*, 1993; Magill *et al.*, 2000; McNulty *et al.*, 1996; Mitchell *et al.*, 2001a; Nadelhoffer *et al.*, 1995). These differences depend on the initial N status of the site and the rate at which sites progress toward saturation (Aber *et al.*, 1998). The heterogeneous nature of forest ecosystems, and the combined effects of factors (e.g. land-use history, forest cover, and hydrologic flow paths) have made it difficult to predict vulnerabilities to high N deposition within and across regions. Factors such as climate (Mitchell *et al.*, 1996; Murdoch *et al.*, 1998) and disturbance create further complexity, especially for temporal patterns of N loss in drainage waters (Aber *et al.*, 2002).

Small watersheds have long been recognized as a useful tool for investigating how ecosystems respond to changes caused by both natural and human perturbations (Bormann and Likens, 1979; Church, 1997; Likens and Bormann, 1995). Provided that loss to groundwater is negligible, watershed N accumulation or loss can be determined by subtracting outputs in streamflow from inputs from atmospheric deposition. This approach assumes that there is no source of N via mineral weathering and no significant gains or losses of N through gaseous exchange with the atmosphere. Mineralogical sources of N can contribute to N losses in some areas of the United States (Holloway and Dahlgren, 1999; Holloway *et al.*, 1998), but are not an important source of N in watersheds of the northeastern United States. Nitrogen budgets may also be affected by N fixation and denitrification; however in our study watersheds, these gains or losses are thought to be negligible compared to fluxes through hydrologic pathways (Bormann *et al.*, 1993; Bormann and Likens, 1979; Bowden *et al.*, 1990; Bowden, 1986). Annual efflux of nitrogenous gases is minor (<0.1 to 1.5 kg N ha^{-1} yr^{-1}) in relatively undisturbed, forest watersheds of the northeastern United States (Ashby *et al.*, 1998; Bowden *et al.*, 1990; Bowden, 1986). However, it is difficult to measure gaseous N flux at the small watershed scale because of the large spatial variability within watersheds (Bohlen *et al*., 2000) and problems associated with measurement methodology (Bowden *et al*., 1990).

Results from individual watershed studies have provided data on N retention and loss in the northeastern United States; however, there have been few attempts to synthesize these data to examine regional patterns. Several studies have compared streamwater concentrations of N (Hornbeck *et al*., 1997; Stoddard, 1994), but these analyses do not include stream discharge data necessary to calculate fluxes. Past watershed N budget comparisons that have been conducted are limited to a small number of watersheds (Campbell *et al*., 2000) or target specific areas within the northeastern region of the United States (Goodale *et al*., 2000; Lovett *et al*., 2000).

In North America, the concern over N saturation has largely focused on the northeastern United States because this area receives some of the greatest amounts of N deposition in North America (Clarke *et al*., 1997; Munger and Eisenreich, 1983). To examine N input-output budgets in this region, data were compiled for 24 relatively small, forest watersheds (Table I). The objectives of this analysis were to establish ranges for fluxes of inorganic N (NH_4-N and NO_3-N) in precipitation and streamwater and to determine if there are general spatial patterns in N retention across the region. Data from the 11 most intensively studied watersheds were used to determine if there were general relationships between N retention and watershed characteristics. Forest cover, hydrology, soil properties, and disturbance history were examined as possible controls on N cycling. These analyses will enable researchers to put results of individual watershed studies in a regional context and will provide a better understanding of how watersheds differ in their capacity to retain N. Furthermore, it will improve our ability to predict N export and will help identify areas that may be sensitive to conditions of N saturation.

2. Methods

Watersheds were chosen based on size, land-use, and sampling interval. Only small (<1000 ha), forest watersheds that were free of recent (at least 50 yr), large-scale physical disturbance were considered. These criteria eliminated local differences in N export related to deforested or developed land. Also, only watersheds with sampling intervals of three weeks or less were chosen to ensure that seasonal patterns and higher flow events were adequately represented. We identified 24 watersheds from 15 sites throughout the region that met the aforementioned criteria. These sites covered an area from 39 to 46°N latitude and 68 to 80°W longitude. Details about watershed characteristics and sampling procedures are given in Table I.

Annual inorganic N budgets (NH_4-N and NO_3-N) were compiled for each watershed using stream and precipitation volume and chemistry data. A 1 June water

TABLE I

Watershed characteristics and sampling regime

Site	State	Abbrev.	Agency collecting data	Reference	Latitude, longitude	Name of NADP site (code)	Distance to NADP site (km)	Elevation of NADP site (m)	Stream
Cockaponset	CT	CT	USDA Forest Service	Hornbeck *et al*. (1990)	41°24′N, 72°32′W	Stilwell Lake (NY51)	127	190	Unnamed stream
						West Point (NY99)	127	200	
Acadia	ME	ACC	Univ. of Maine	Nelson (2002)	44°21′N, 68°13′W	Acadia Natl. Park (ME98)	On-site	130	Cadillac Brook
		ACH			44°20′N, 68°17′W				Hadlock Brook
Bear Brook		EBB[a]	Univ. of Maine	Norton and Fernandez (1999)	44°52′N, 68°06′W	–	–	–	East Bear Brook
Weymouth Point		WPT	USDA Forest Service	Hornbeck *et al*. (1990)	45°56′N, 69°17′W	Greenville Stn. (ME09)	57	320	Unnamed stream
Unnamed Tributary to Herrington Creek	MD	HCR	Univ. of Maryland	Castro and Morgan (2000)	39°28′N, 79°26′W	–	–	–	Unnamed tributary to Herrington Creek
Bowl	NH	BE	Syracuse Univ.	Martin *et al*. (2000)	43°56′N, 71°23′W	Hubbard Brook (NH02)	26	250	East Branch
		BW			43°56′N, 71°24′W				West Branch
		BU			43°56′N, 71°24′W				Upper Branch
		BL			43°56′N, 71°23′W				Lower Branch
Cone Pond		CP[a]	USDA Forest Service	Hornbeck *et al*. (1997)	43°54′N, 71°36′W	Hubbard Brook (NH02)	10	250	Cone Pond Inlet
Hubbard Brook		HB6[a]	Inst. Ecosystem Studies	Likens and Bormann (1995)	43°57′N, 71°44′W	Hubbard Brook (NH02)	On-site	250	Watershed 6
		HB9[a]			43°55′N, 71°45′W				Watershed 9
Mt. Success		MTS	USDA Forest Service	Hornbeck *et al*. (1990)	44°30′N, 71°03′W	Hubbard Brook (NH02)	81	250	Unnamed stream
Biscuit Brook	NY	BSB[a]	US Geological Survey	Murdoch and Stoddard (1992)	41°59′N, 74°30′W	Biscuit Brook (NY68)	On-site	630	Biscuit Brook
Huntington		HW[a]	SUNY-ESF	Mitchell *et al*. (2001b)	44°00′N, 74°13′W	Huntington Wildlife (NY20)	On-site	500	Archer Creek
Leading Ridge	PA	LR[a]	Pennsylvania State Univ.	Lynch and Corbett (1989)	40°44′N, 77°55′W	Leading Ridge (PA42)	On-site	290	Watershed 1
Lye Brook	VT	LB4	USDA Forest Service	Campbell *et al*. (2002)	43°07′N, 73°03′W	Bennington (VT01)	29	310	Watershed 4
		LB6			43°07′N, 73°02′W				Watershed 6
		LB8			43°07′N, 73°02′W				Watershed 8
Sleepers River		SR[a]	US Geological Survey	Hornbeck *et al*. (1997)	44°29′N, 72°10′W	Underhill (VT99)	63	400	Watershed 9
Fernow	WV	F4[a]	USDA Forest Service	Edwards and Helvey (1991)	39°03′N, 79°41′W	Parsons (WV18)	On-site	510	Watershed 4
		F10[a]			39°03′N, 79°41′W				Watershed 10
		F13[a]			39°03′N, 79°41′W				Watershed 13

[a] Intensively monitored site.

[b] Calculated as the mean of the maximum and minimum elevation.

TABLE I
(continued)

Site	State	Abbrev.	Water years start/stop	Total years	Sampling interval (wks.)	Streamflow measurement	Watershed area (ha)	Elevation[b] (m)	Vegetation	Soil classification
Cockaponset	CT	CT	1980–1981/1984–1985	5	1–3	Modeled	7	140	Central hardwoods	Typic Dystrochrept, Aeric Haplaquept
Acadia	ME	ACC	1999–2000	1	1–2	Stream contour	32	300	Mixed northern hardwoods/spruce-fir	Typic Haplorthod
		ACH	1999–2000	1	1–2	Stream contour	47	260	Spruce-fir	Typic Haplorthod
Bear Brook		EBB[a]	1989–1990/1998–1999	10	1	V-notch weir	11	370	Northern hardwoods	Typic Haplorthod
Weymouth Point		WPT	1980–1981/1986–1987	7	1–3	Modeled	72	300	Spruce-fir	Aquic Haplorthod, Aeric Haplaquept
Unnamed tributary to Herrington Creek	MD	HCR	1996–1997	1	1	Stream contour	255	770	Central hardwoods	Typic Dystrochrept, Hapludult, Fragiaquult
Bowl	NH	BE	1995–1996/1996–1997	2	2	Modeled	271	840	Northern hardwoods	Typic Haplorthod
		BW	1995–1996/1996–1997	2	2	Modeled	394	860	Northern hardwoods	Typic Haplorthod
		BU	1995–1996/1996–1997	2	2	Modeled	15	760	Northern hardwoods	Typic Haplorthod
		BL	1995–1996/1996–1997	2	2	Modeled	31	750	Northern hardwoods	Typic Haplorthod
Cone Pond		CP[a]	1990–1991/1997–1998	8	1	V-notch weir	33	570	Mixed northern hardwoods/spruce-fir	Typic and Lithic Haplorthod
Hubbard Brook		HB6[a]	1979–1980/1997–1998	19	1	V-notch weir, flume	13	670	Northern hardwoods	Typic Haplorthod
		HB9[a]	1995–1996/1997–1998	3	1	V-notch weir	68	790	Mixed northern hardwoods/spruce-fir	Typic Haplorthod
Mt. Success		MTS	1979–1980/1980–1981	2	1–3	Modeled	18	500	Northern hardwoods	Typic Fragiorthod
Biscuit Brook	NY	BSB[a]	1984–1985/1996–1997	13	1	V-notch weir	990	880	Northern hardwoods	Typic and Lithic Dystrochrept
Huntington		HW[a]	1995–1996/1997–1998	3	1	H flume	135	640	Northern hardwoods	Typic Haplorthod
Leading Ridge	PA	LR[a]	1979–1980/1992–1993	14	1	V-notch weir	123	380	Central hardwoods	Typic Dystrochrept, Typic and Aquic Fragiudult
Lye Brook	VT	LB4	1994–1995	1	2	Modeled	163	570	Mixed northern hardwoods/spruce-fir	Typic and Lithic Haplorthod
		LB6	1994–1995	1	2	Modeled	106	710	Mixed northern hardwoods/spruce-fir	Typic and Aquic Haplorthod
		LB8	1994–1995	1	2	Modeled	130	790	Mixed northern hardwoods/spruce-fir	Typic Humaquept, Epiaquod, Haplorthod
Sleepers River		SR[a]	1992–1993/1996–1997	5	1	V-notch weir	41	600	Northern hardwoods	Typic Dystrochrept
Fernow	WV	F4[a]	1989–1990/1997–1998	9	1	V-notch weir	39	800	Central hardwoods	Typic Dystrochrept
		F10[a]	1989–1990/1996–1997	8	1	H flume	15	760	Central hardwoods	Typic Dystrochrept
		F13[a]	1989–1990/1996–1997	8	1	H flume	14	760	Central hardwoods	Typic Dystrochrept

year (e.g. WY 1992–1993 is from 1 June 1992 through 31 May 1993) was used to calculate annual fluxes because this period usually provides the best correlation between annual precipitation and streamflow (Likens and Bormann, 1995). To determine watershed inputs, we used weekly precipitation chemistry data from the National Atmospheric Deposition Program (NADP, 2002) except at Bear Brook and the unnamed tributary to Herrington Creek where wet deposition data were collected independent of the NADP program. Wet deposition measurements at these sites are comparable to measurements from NADP sites since the equipment and methods used to collect data are nearly identical (Castro and Morgan, 2000; Kahl *et al.*, 1999). Samples collected as part of the NADP program are sent to a central laboratory at the University of Illinois at Urbana-Champaign and are analyzed for NO_3^- using an ion chromatograph and NH_4^+ using a flow injection analyzer. A thorough description of NADP sampling and analytical procedures is available through the NADP program office (NADP, 2002). For each NADP site, monthly input values were calculated by summing the product of weekly precipitation volume and chemical concentrations. In cases where there were insufficient data to characterize a monthly summary period (NADP, 2002), we used long-term monthly means based on all the data available since the inception of the NADP program in WY 1979–1980. Of the 15 sites included in this study, six had NADP collectors located on-site (Table I). For watersheds that lacked on-site NADP collectors, N concentrations in precipitation were based on data from the closest NADP site. At Cockaponset, data from two NADP sites were used because the closest NADP site (Stilwell Lake, NY) was discontinued in 1984.

Use of these NADP data assumes that N concentrations in precipitation at the closest NADP site were representative of N concentrations at corresponding watersheds. The NADP collectors were located within 130 km of the watersheds and the difference in elevation between NADP collectors and the midpoint elevation of watersheds was <610 m (Table I). These differences in distance and elevation, as well as differences in landscape features such as vegetation type, forest gaps, and aspect, may affect estimates of N deposition (Weathers *et al.*, 2000). However, concentrations of N in precipitation are fairly uniform across these sites (range in NH_4-N = 0.1 to 0.2 mg L^{-1}; range in NO_3-N = 0.2 to 0.5 mg L^{-1}) and our data, as well as data from other studies (Ito *et al.*, 2002; Lovett and Kinsman, 1990; Miller *et al.*, 1993; Ollinger *et al.*, 1995), indicate that concentrations of N in precipitation are not related to elevation. There are spatial trends in concentrations of N in precipitation across the region. Ollinger *et al.* (1995) found that in the northeastern United States, NO_3-N was significantly related to longitude, and both latitude and longitude were significant predictors of NH_4-N. However, differences in the concentrations of N in precipitation over a distance of less than 130 km are minor.

Since N inputs are influenced more by the quantity of precipitation than by concentrations of N, precipitation measurements were obtained from the closest precipitation gage associated with each watershed. At Acadia National Park, Huntington and Leading Ridge, we used precipitation volume measured as part of the NADP program to calculate budgets. For all other sites, precipitation measurements were obtained from closer rain gages operated independent of the NADP program. The only sites that did not have on-site precipitation collectors were Lye Brook and the Bowl. For these sites, precipitation volume measurements were based on data from nearby (<10 km) National Weather Service (NWS) stations (Dorset, VT and Tamworth, NH, respectively) that were corrected for elevation using regression equations developed for each month of the year (Ollinger *et al.*, 1995).

Dry deposition was not included in this analysis due to the paucity of data available, and uncertainty associated with its measurement. In a regional deposition model for the northeastern United States, Ollinger *et al.* (1995) determined that dry N deposition (measured as the sum of gaseous HNO_3-N and particulate NO_3-N and NH_4-N) was approximately 20–46% of wet N deposition. At Fernow, Hubbard Brook and Lye Brook, dry deposition data are measured on-site as part of the Clean Air Status and Trends Network (US Environmental Protection Agency, Washington DC). Mean annual dry N deposition at these sites was respectively 2.1, 0.4, and 2.6 kg N ha^{-1} yr^{-1} (6–36% of wet N deposition).

Cloud and fog water inputs were also not included in this analysis. Several studies have shown that N deposited in cloud and fog water can be important at high elevation sites in the northeastern United States, such as Whiteface Mountain in New York ($\sim$6–7 kg N ha^{-1} yr^{-1} at 1000 m) (Lovett and Lindberg, 1993; Miller *et al.*, 1993). However, at lower elevation sites, such as the Huntington Forest (Lovett and Lindberg, 1993) and Hubbard Brook (Weathers *et al.*, 1988), N inputs in cloud and fog water are negligible. Since the N inputs reported in this study were based solely on wet deposition, and do not include dry deposition or deposition from fog and cloud water, they under-represent the total N atmospheric inputs.

Streamwater outputs were obtained from independent monitoring programs at each watershed. The studies spanned different periods (1 to 19 yr) and typically used different protocols for sample collection and analysis (Table I). At each stream, samples were collected at specified intervals (Table I) and were analyzed for NH_4-N and NO_3-N. Streamwater outputs of N were calculated by multiplying mean concentrations by corresponding water fluxes. At 14 of the watersheds, streamflow was measured using stage-height recorders and stream-channel controls including weirs, flumes, or natural stream contours. At the other 10 watersheds streamflow was estimated using the BROOK90 hydrological model (Federer, 1997; Federer and Lash, 1978) (Table I).

BROOK90 is a lumped-parameter model that can be used to estimate streamflow for small, forested watersheds. The model simulates vertical water movement at a single point, and consequently works best for fairly uniform watersheds, such as those included in this study. BROOK90 requires daily precipitation, and minimum and maximum air temperature input variables. The model was run on a daily time step to predict streamflow expressed as mm day^{-1}. For those watersheds where streamflow was measured directly, evapotranspiration was calculated on a water year basis as the difference between precipitation inputs and stream discharge. For those sites where the hydrology was simulated with BROOK90, evapotranspiration was calculated by the model.

Export calculations differed according to the methods established at each watershed (Table I). Measurements of solute export can be influenced by the frequency of data collection, particularly for elements that are well correlated with streamflow (Swistock *et al.*, 1997). Intermittent stream sampling generally characterizes low flow better than event flow because there is a greater likelihood that samples will be collected during the more common, low flow period. For NO_3-N and NH_4-N, this should not result in substantial errors in the export calculation of N since these solutes generally do not exhibit large responses to streamflow on an annual basis (Swistock *et al.*, 1997). However, greater sampling frequency, event sampling, and measured (rather than modeled) streamflow yield the best estimates of N output.

The dissolved organic fraction of N (DON) was not included in the N budgets for these watersheds. At some of the watersheds, DON is measured in precipitation (independent of the NADP program) and more commonly in streamflow, but the data have only been collected recently and analytical procedures vary among studies making comparisons difficult. Although a significant fraction of N exports may be comprised of DON (Campbell *et al.*, 2000; Goodale *et al.*, 2000; McHale *et al.*, 2000), a study of eight watersheds in Vermont and New Hampshire found that the net difference between DON inputs and outputs did not exceed 1.5 kg N ha^{-1} yr^{-1} (Campbell *et al.*, 2000).

Of the 24 total watersheds included in the present study, 11 watersheds from 8 sites were selected for more detailed analyses (Biscuit Brook, Bear Brook, Cone Pond, Fernow, Hubbard Brook, Huntington, Leading Ridge, Sleepers River). Watersheds used in the detailed analysis were selected using more stringent criteria, which included: continuous streamflow measurement, weekly chemical sampling, and long-term records (>2 yr). We were not able to compare data for the same years at all sites because the collection periods varied in length and did not always coincide. Data collected before WY 1979–1980 were not used in this analysis because NADP data were not available before this time and because we wanted to analyze more recent patterns in N deposition and streamwater. At each watershed, mean annual input and output values were calculated using all the data that were available since WY 1979–1980. Data after 1997–1998 were not included because of the disturbance effects of a widespread ice storm that occurred in the region in January 1998. Budgets were developed by subtracting outputs from inputs.

TABLE II

Mean annual watershed hydrological budgets (mm ha^{-1} yr^{-1}) for all the years available from WY 1979–1980 through WY 1997–1998. Evapotranspiration is calculated as precipitation minus streamflow. Streamflow and evapotranspiration are also expressed as a percentage of total precipitation

Site	State	Abbrev.	Precipitation (mm)	Streamflow (mm)	Evapotranspiration (mm)	Streamflow (%)	Evapotranspiration (%)
Cockaponset	CT	CT	1350	790	560	59	41
Acadia	ME	ACC	1440	920	520	64	36
		ACH	1440	1080	360	75	25
Bear Brook		EBB[a]	1250	920	330	74	26
Weymouth Point		WPT	970	290	680	30	70
Unnamed Tributary to Herrington Creek	MD	HCR	1430	940	490	66	34
Bowl	NH	BE	1930	1370	560	71	29
		BW	1960	1360	600	69	31
		BU	1860	1310	550	70	30
		BL	1860	1300	560	70	30
Cone Pond		CP[a]	1280	670	610	52	48
Hubbard Brook		HB6[a]	1420	900	520	63	37
		HB9[a]	1630	1070	560	66	34
Mt. Success		MTS	900	470	430	52	48
Biscuit Brook	NY	BSB[a]	1520	970	550	64	36
Huntington		HW[a]	1210	830	380	69	31
Leading Ridge	PA	LR[a]	1050	470	580	45	55
Lye Brook	VT	LB4	1240	600	640	48	52
		LB6	1330	720	610	54	46
		LB8	1390	740	650	53	47
Sleepers River		SR[a]	1320	740	580	56	44
Fernow	WV	F4[a]	1460	710	750	49	51
		F10[a]	1450	690	760	48	52
		F13[a]	1450	890	560	61	39

[a] Intensively monitored site.

3. Results and Discussion

3.1. Water budgets

Annual average precipitation ranged from 900 mm at Mt. Success to 1960 mm at the West Branch of the Bowl (Table II). Annual average streamflow ranged from 290 mm at Weymouth Point to 1370 mm at the East Branch of the Bowl, and was

30 to 75% of precipitation. Annual average evapotranspiration ranged from 330 to 760 mm and was 25 to 70% of precipitation. The relatively large range in measurements of precipitation, streamflow, and evapotranspiration may be partially due to the short sampling period at some sites. However, the range was fairly wide even among watersheds with relatively long hydrological records (e.g., Biscuit Brook, East Bear Brook, Hubbard Brook Watershed 6, and Leading Ridge).

3.2. Ammonium

Streamwater NH_4-N outputs were low and NH_4-N inputs in precipitation were consistently greater than streamwater outputs at all watersheds. The relatively small outputs of NH_4-N indicate that nearly all the NH_4-N added in precipitation is being retained or transformed within these watersheds (Table III). Concentrations of NH_4-N in precipitation ranged from 0.1 to 0.2 mg L^{-1} and fluxes ranged from 0.9 to 2.8 kg N ha^{-1} yr^{-1}. In comparison, streamwater concentrations (<0.1 mg L^{-1}) and fluxes (<0.2 kg N ha^{-1} yr^{-1}) were markedly lower. Annual contributions of NH_4-N to the DIN retained in forest watersheds ranged from 0.7 to 2.7 kg N ha^{-1} yr^{-1} (26–92%). Possible transformations that could cause low NH_4-N outputs include uptake by vegetation, microbial immobilization and nitrification, and adsorption on soil surfaces.

3.3. Nitrate

Concentrations of NO_3-N in precipitation ranged from 0.2 to 0.5 mg L^{-1} and fluxes ranged from 1.8 to 5.5 kg N ha^{-1} yr^{-1} (Table III). Streamwater concentrations (<0.1 to 0.8 mg L^{-1}) and fluxes (<0.1 to 5.7 kg N ha^{-1} yr^{-1}) were generally lower than concentrations and fluxes in precipitation. However unlike NH_4-N, there was a large range in streamwater NO_3-N exports, indicating large differences in the source, generation and processing of NO_3-N among watersheds. All watersheds retained NO_3-N on an annual basis (0.1 to 5.0 kg N ha^{-1} yr^{-1}) except for Watershed 4 at the Fernow Experimental Forest, which had a net loss of 0.7 kg N ha^{-1} yr^{-1}. Since high leaching loss of NO_3-N is considered to be a sign that N inputs exceed the biological demand for N, it has been suggested that this watershed may be experiencing N saturation (Peterjohn *et al.*, 1996). All other watersheds accumulated NO_3-N, although in some cases the differences between inputs and outputs were relatively low, such as Mt. Success (0.1 kg N ha^{-1} yr^{-1}).

3.4. Dissolved inorganic N

DIN (NH_4-N + NO_3-N) budgets show that at all watersheds, precipitation inputs of DIN exceeded outputs resulting in a net DIN accumulation of 1.2 to 7.3 kg N ha^{-1} yr^{-1} (Table III). The range in DIN inputs was 2.7 to 8.1 kg N ha^{-1} yr^{-1} (mean = 6.4 kg N ha^{-1} yr^{-1}; median = 7.0 kg N ha^{-1} yr^{-1}). Outputs of DIN ranged from 0.1 to 5.7 kg N ha^{-1} yr^{-1} (mean = 2.0 kg N ha^{-1} yr^{-1}; median

TABLE III

Mean annual watershed NH_4-N, NO_3-N, and DIN (NH_4-N + NO_3-N) budgets (kg N ha^{-1} yr^{-1}) for all the years available from WY 1979–1980 through WY 1997–1998. Total N retention/loss is calculated as inputs minus outputs. Total percent DIN retention is calculated using the equation ([input-output]/input). Values in parentheses indicate respective percentage of NH_4-N and NO_3-N in DIN retained

Site	State	Abbrev.	Inputs			Outputs			Total N retention/loss			Total DIN
			NH_4-N	NO_3-N	DIN	NH_4-N	NO_3-N	DIN	NH_4-N (%)	NO_3-N (%)	DIN	retention (%)
			(kg N ha^{-1} yr^{-1})									
Cockaponset	CT	CT	1.9	5.0	6.9	0.1	<0.1	0.1	1.8 (26)	5.0 (74)	6.8	99
Acadia	ME	ACC	1.5	3.0	4.5	0.1	0.1	0.2	1.4 (33)	2.9 (67)	4.3	96
		ACH	1.5	3.0	4.5	0.1	1.2	1.3	1.4 (44)	1.8 (56)	3.2	71
Bear Brook		EBB[a]	1.3	2.5	3.8	<0.1	0.6	0.6	1.3 (41)	1.9 (59)	3.2	84
Weymouth Point		WPT	0.9	1.8	2.7	0.2	0.2	0.4	0.7 (30)	1.6 (70)	2.3	85
Unnamed Tributary to Herrington Creek	MD	HCR	2.4	4.4	6.8	0.1	2.2	2.3	2.3 (51)	2.2 (49)	4.5	66
Bowl	NH	BE	2.2	5.4	7.6	0.2	2.8	3.0	2.0 (43)	2.6 (57)	4.6	61
		BW	2.3	5.5	7.8	0.2	2.5	2.7	2.1 (41)	3.0 (9)	5.1	65
		BU	2.2	5.2	7.4	0.1	2.7	2.8	2.1 (46)	2.5 (54)	4.6	62
		BL	2.1	5.2	7.3	0.2	2.9	3.1	1.9 (45)	2.3 (55)	4.2	58

[a] Intensively monitored site.
[b] Value is based on one water year (1992–1993).

TABLE III
(continued)

Site	State	Abbrev.	Inputs			Outputs			Total N retention/loss			Total DIN
			NH_4-N	NO_3-N	DIN	NH_4-N	NO_3-N	DIN	NH_4-N (%)	NO_3-N (%)	DIN	retention (%)
			(kg N ha^{-1} yr^{-1})									
Cone Pond		CP[a]	1.7	3.8	5.5	0.2	<0.1	0.2	1.5 (28)	3.8 (72)	5.3	96
Hubbard Brook		HB6[a]	1.8	4.3	6.1	0.1	1.2	1.3	1.7 (35)	3.1 (65)	4.8	79
		HB9[a]	2.0	4.6	6.6	0.1	0.4	0.5	1.9 (31)	4.2 (69)	6.1	92
Mt. Success		MTS	1.1	3.3	4.4	<0.1	3.2	3.2	1.1 (92)	0.1 (8)	1.2	27
Biscuit Brook	NY	BSB[a]	2.4	5.0	7.4	0.1[b]	4.0	4.1	2.3 (70)	1.0 (30)	3.3	45
Huntington		HW[a]	1.6	3.4	5.0	0.2	2.7	2.9	1.4 (67)	0.7 (33)	2.1	42
Leading Ridge	PA	LR[a]	2.4	4.7	7.1	–	<0.1	–	–	4.7	–	–
Lye Brook	VT	LB4	2.5	4.7	7.2	<0.1	1.0	1.0	2.5 (40)	3.7 (60)	6.2	86
		LB6	2.7	5.1	7.8	0.1	2.5	2.6	2.6 (50)	2.6 (50)	5.2	67
		LB8	2.8	5.3	8.1	0.1	0.7	0.8	2.7 (37)	4.6 (63)	7.3	90
Sleepers River		SR[a]	2.4	4.2	6.6	0.1	1.6	1.7	2.3 (47)	2.6 (53)	4.9	74
Fernow	WV	F4[a]	2.5	5.0	7.5	<0.1	5.7	5.7	2.5	–0.7	1.8	24
		F10[a]	2.5	5.0	7.5	<0.1	1.1	1.1	2.5 (39)	3.9 (61)	6.4	85
		F13[a]	2.5	5.0	7.5	<0.1	4.2	4.2	2.5 (76)	0.8 (24)	3.3	44

[a] Intensively monitored site.
[b] Value is based on one water year (1992–1993).

= 1.7 kg N ha^{-1} yr^{-1}). Percent N watershed retention ranged from 24 to almost 100% (mean = 69%; median = 71%). In some cases, such as Cockaponset, Cone Pond, and Cadillac Brook at Acadia, nearly all the wet N deposition was retained within the watershed (99, 96, and 96%, respectively). Other watersheds, such as Watershed 4 at the Fernow Experimental Forest and Mt Success, retained much less of the annual wet N input (24 and 27% respectively). At all watersheds, NO_3-N constituted a greater proportion of DIN inputs compared to NH_4-N. Results for streamwater were similar, with NO_3-N constituting a greater proportion of DIN outputs compared to NH_4-N at most watersheds. The only watersheds where NH_4-N outputs were greater than or equal to NO_3-N outputs were Cockaponset, Cone Pond, Cadillac Brook at Acadia, and Weymouth Point. These data indicate that NO_3-N is typically the dominant form of inorganic N in both precipitation and streamwater, and that NH_4-N is lower in precipitation and near zero in streamwater.

3.5. Regional patterns

The watersheds we examined occur along a gradient of atmospheric N deposition, so spatial patterns in N retention among watersheds of the region were assessed. In the northeastern United States, the greatest N deposition occurs in Pennsylvania, New York, western Maryland and northern West Virginia (NADP, 2002). In our study, the lowest wet N inputs were found at the inland sites in Maine (East Bear Brook and Weymouth Point), which are at the extreme northeast portion of the study region. These sites have lower N concentrations and receive less rainfall (due to lower elevation), and consequently have lower N inputs. Wet deposition of N at the other watersheds did not exhibit distinct spatial patterns (e.g. gradients of increasing N deposition toward emission sources in the midwestern United States). At these watersheds, differences in atmospheric concentrations of N may be small or local factors that affect precipitation volume (e.g. elevation) may confound regional spatial relationships.

There were no apparent regional patterns in streamwater exports of N. Fluxes were highly variable even among adjacent watersheds that had similar characteristics and N loading. The large range in stream N exports, compared to the more narrow range in precipitation inputs, indicates differences in N cycling within watersheds. A portion of the variability in stream N outputs may also be attributed to differences in sampling procedures as well as to the duration of each study (Table I). Use of NADP data eliminated potential problems because of site differences in chemical techniques and the calculation of wet N inputs.

3.6. Analysis of more intensively monitored sites

To address some concerns that may be associated with sampling at several sites, eleven watersheds (located at East Bear Brook, Biscuit Brook, Cone Pond, Fernow, Hubbard Brook, Huntington, Leading Ridge, Sleepers River) with more intensive long-term monitoring programs were investigated beyond the analysis of the larger

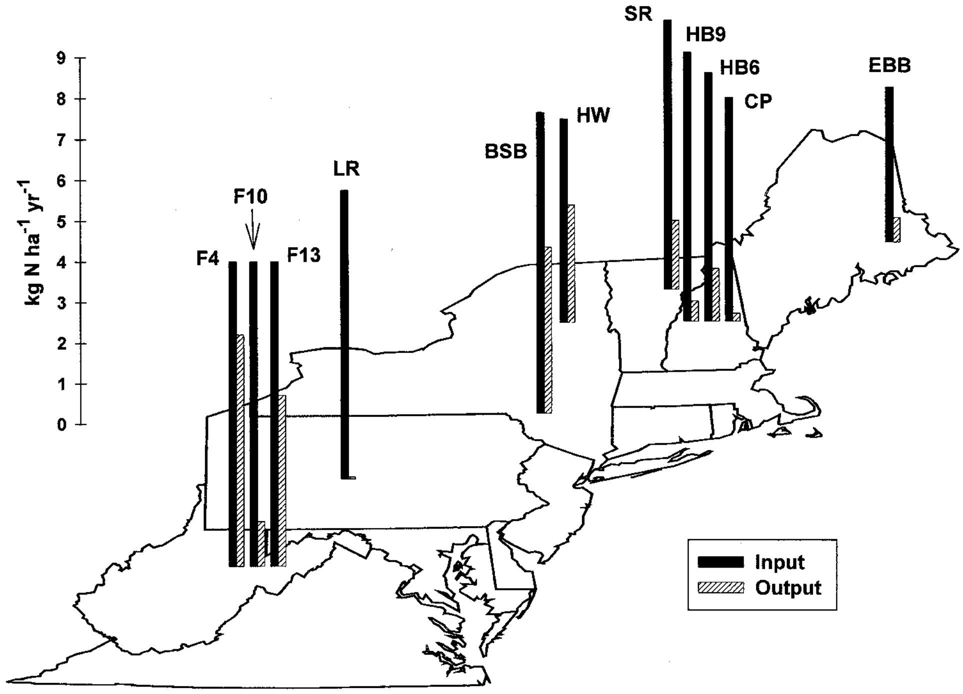

Figure 1. Inputs of DIN in wet-only precipitation and outputs of DIN in streamwater (kg N ha^{-1} yr^{-1}) at the more intensively monitored study watersheds. Output data for Leading Ridge (LR) do not include NH_4-N values because NH_4-N was not measured in streamwater at this site.

data set. For these watersheds, wet DIN inputs in precipitation ranged from 3.8 to 7.5 kg N ha^{-1} yr^{-1} and stream outputs ranged from 0.2 to 5.7 kg N ha^{-1} yr^{-1} (Figure 1, Table III). For these intensively monitored sites, there was still a large range in percent DIN retention (24 to 96%).

3.7. Factors affecting N retention

One of the main objectives of our analysis was to examine factors that affect N retention in forest ecosystems. Since hydrological values are used to calculate fluxes, factors that affect precipitation or streamflow volume can also affect inputs and outputs of N. There was a significant relationship between mean annual streamflow and precipitation (streamflow (mm) = 0.72 × precipitation (mm) – 181.25; r^2 = 0.49; $P < 0.02$) at the intensively monitored sites indicating that streamflow is primarily affected by the amount of precipitation falling on a watershed rather than other factors such as differences in flow paths and vegetation.

The large range in precipitation among sites partially arises from the range in watershed elevation. The mid-point elevation of the intensively monitored watersheds (calculated as the mean of the maximum and minimum watershed elevation) ranged from 370 m at East Bear Brook, to 880 m at Biscuit Brook. There was a significant relationship between precipitation and elevation at these watersheds (precipitation (mm) = 0.84 $\times$ elevation (m) + 816.52; r^2 = 0.74; $P < 0.001$) showing that high-elevation watersheds typically received the greatest amount of precipitation. This relationship is primarily due to orographic effects and is consistent with similar studies in the northeastern United States (Dingman *et al*., 1988; Lovett and Kinsman, 1990). The relationship between wet DIN deposition and elevation was also significant (N inputs (kg N ha^{-1} yr^{-1}) = 0.005 $\times$ elevation (m) + 3.43; r^2 = 0.39; $P < 0.04$) indicating that the higher elevation sites included in this study also receive higher wet N deposition.

In a synthesis of N watershed budgets in Europe, Dise and Wright (1995) found that bulk inputs of inorganic N in precipitation were the most important predictor of N exports in streamwater of 41 variables examined (N outputs (kg N ha^{-1} yr^{-1}) = 0.48 $\times$ N inputs (kg N ha^{-1} yr^{-1}) – 2.17; r^2 = 0.69; $P < 0.001$). However, at European watersheds with N inputs of less than 10 kg N ha^{-1} yr^{-1}, nearly all the N was retained and most of the significant leaching was found at watersheds receiving inputs greater than 25 kg N ha^{-1} yr^{-1}. There was not a significant relationship between wet DIN inputs and stream outputs for the intensively monitored watersheds in our study, presumably because deposition of N is much lower in the northeastern United States compared to Europe. At some of the European sites bulk N inputs exceeded 60 kg N ha^{-1} yr^{-1}. The threshold of 25 kg N ha^{-1} yr^{-1} exceeds even the highest wet N inputs (8.1 kg N ha^{-1} yr^{-1}) of the watersheds in our study. Differences between bulk deposition and wet deposition are not nearly enough to account for this discrepancy and estimates of total N deposition in the northeastern United States (wet and dry) are thought to be less than 12 kg N ha^{-1} yr^{-1} (Ollinger *et al*., 1995).

3.8. Factors controlling N retention

Complex processes that occur within watersheds regulate N export. Site characteristics, such as hydrology, forest cover, and land-use, largely influence these interactions and further complicate assessment of the relationship between N inputs and outputs.

3.8.1. *Influence of Hydrology*

Hydrologic flow paths can be a major factor influencing N retention and temporal patterns of stream N loss in forest watersheds (Mitchell, 2001). Watersheds with thin or porous soils and high infiltration rates have less capacity to retain N (Lajtha *et al*., 1995). Similarly, numerous studies have shown that NO_3^- rapidly leaches through soils to streams during snowmelt runoff (Galloway *et al*., 1987; Rascher *et al*., 1987) and high flow events (Wigington *et al*., 1996). The relationship between discharge and N export is typically stronger during the dormant season when biotic retention of N is lower.

At Biscuit Brook, Murdoch and Stoddard (1992) observed increases in NO_3^- concentrations with increasing discharge throughout most of the year, and relatively high N stream losses during the growing season. Several hypotheses have been proposed to explain the high exports of NO_3-N (4.0 kg N ha^{-1} yr^{-1}) at Biscuit Brook. Burns *et al*. (1998) suggested that high streamwater NO_3-N concentrations that occur throughout the growing season are the result of a deep groundwater source of NO_3-N. Groundwater in this watershed is recharged with NO_3-N during the fall and early spring. This groundwater provides NO_3-N to surface waters during base flow in summer. Since NO_3-N in deep groundwater is affected by reduced biotic activity, concentrations of NO_3-N remain relatively high throughout the growing season.

Lovett *et al*. (2000) reasoned that if groundwater sources of NO_3-N drive streamwater NO_3-N concentrations, a relationship between Ca^{2+} and NO_3-N would be expected since Ca^{2+} concentrations in groundwater are high due to greater contact with less-weathered bedrock and deep till. However, there was only a strong relationship between NO_3-N and Ca^{2+} at high NO_3-N streams during the winter, while during the summer this relationship was not evident. This pattern suggests that the relationship between NO_3-N and Ca^{2+} is due to NO_3-N induced leaching of Ca^{2+} and is not indicative of a groundwater source of NO_3-N. Furthermore, Lovett *et al*. (2000) found a poor relationship between NO_3-N concentrations and physical features of Catskill watersheds that might be expected to affect hydrologic residence times. They concluded that hydrologic differences are probably not driving differences in NO_3-N concentrations among watersheds, and hypothesized that among-watershed differences in tree species composition and historical land-use patterns described in the following sections are more likely to explain spatial patterns of N export and retention in the Catskill Mountains. This conclusion contrasts to the findings of Creed and Band (1998), working within a series of watersheds in Canada with more uniform vegetation than the Catskills. They suggested the importance of topography and hydrological factors in controlling surface water NO_3-N concentrations.

3.8.2. *Influence of Vegetation*

The effect of forest cover on N retention may be due to differences in N uptake and litter quality. Soil C:N ratios have been shown to be good predictors of DIN export in drainage water (Gundersen *et al*., 1998; McNulty *et al*., 1991) and coniferous species typically have higher C:N ratios than deciduous species due to the lower N concentration of litter. Higher C:N ratios generally result in higher N immobilization and hence low N leaching at coniferous sites. However, coniferous species also have a much lower demand for N, which under conditions of high N deposition could contribute to greater leaching losses. The importance of forest cover was evaluated with respect to NH_4-N and NO_3-N outputs, but no clear relationship was evident (Figure 2). This lack of a relationship between forest cover and DIN loss provides further evidence of multiple controls on N retention.

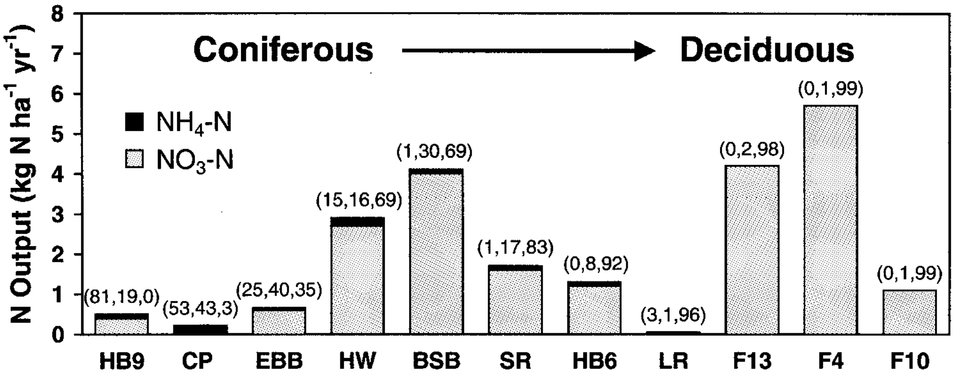

Figure 2. Streamwater DIN outputs (kg N ha^{-1} yr^{-1}) on a gradient from dominant coniferous to dominant deciduous forest cover. Values in parentheses indicate respective percentage of coniferous, mixed, and deciduous forest cover types for each watershed. Data for Leading Ridge (LR) do not include NH_4-N values because NH_4-N was not measured in streamwater at this site.

Despite a poor overall relationship between forest cover and N retention among our study watersheds, other studies have shown that vegetation plays an important role in regulating N losses (Lovett and Rueth, 1999; Magill *et al.*, 2000). The three watersheds of the Fernow Experimental Forest provide an example of how vegetation may influence N retention. The Fernow Experimental Forest is showing signs of N saturation, and is possibly the best case of an N-saturated site in North America (Peterjohn *et al.*, 1996). Several symptoms of N saturation have been identified at the Fernow Experimental Forest including high rates of net nitrification, long-term increases in streamwater concentrations of NO_3-N and base cations, relatively high NO_3-N concentrations in soil solutions, little seasonal variability in streamwater NO_3-N concentrations, and low retention of inorganic NO_3-N compared to other forest watersheds (Peterjohn *et al.*, 1996).

For the Fernow watersheds investigated in our study, Watershed 4 retained only 24% of wet DIN deposition and was the only watershed where mean annual stream NO_3-N outputs exceeded inputs. Watershed 13 had the second highest NO_3-N outputs and retained less than half of wet DIN inputs. In contrast, Watershed 10 had relatively low NO_3-N exports and retained 85% of wet DIN deposition. The three Fernow watersheds have similar climatic and watershed characteristics (e.g. size, elevation, soils, parent material, hydrology, N deposition), and all samples are collected and analyzed using the same methods.

A principal mechanism driving differences in stream N losses at the Fernow Experimental Forest may be related to vegetation. Peterjohn *et al.* (1998) examined N_2O production measured at plots within the boundary of Watershed 4 to evaluate factors that influence susceptibility to N saturation. Differences in N_2O production among plots did not appear to be associated with differences in soil temperature, air temperature, water filled soil pore space, or soil pH. An important factor influencing N_2O production in Watershed 4 appears to be differences in NO_3^- availability associated with tree species composition (Peterjohn *et al*., 1999). Plots with the highest N_2O production were dominated by tree species characterized by low leaf lignin and high soil nitrification rates (e.g. sugar maple (*Acer saccharum*)), presumably due to higher rates of N cycling associated with more rapid litter decomposition. In contrast, plots with low N_2O production were characterized by a greater proportion of species associated with lower rates of soil nitrification (e.g. red oak (*Quercus rubra*) and American beech (*Fagus grandifolia*)). These species-related differences in N retention are consistent with the results for the Catskill streams described in the previous section, as well as those of other studies in the northeastern United States (Lewis and Likens, 2000; Lovett and Rueth, 1999).

It is also possible that the herbaceous layer may influence N retention among Fernow Watersheds. Gilliam *et al*. (2001) found that plots within Watershed 4 with low soil water NO_3^- concentrations were found in areas where lowbush blueberry (*Vaccinium vacillans*) was common. Lowbush blueberry has been shown to acidify the soil, thereby reducing soil N mineralization and nitrification.

The successional status of vegetation may be important in regulating N losses and it has been suggested that aggrading forests may have lower NO_3^- losses because they are thought to have a higher demand for N (Vitousek and Reiners, 1975). Fernow Watershed 4 had a relatively high proportion of old-growth beech and sugar maple (some trees may reach 300 yr old). This stand now appears to be deteriorating rapidly as a result of wind damage, which could contribute to the high NO_3-N losses. Stream export of N from the Bowl may also be affected by the old-growth status of the forest. While N retention at the Bowl (58–65%) was not excessively low compared to some of the other watersheds we investigated, streamwater NO_3^- concentrations tend to be elevated throughout the year, including the growing season, indicating an excess of N (Martin *et al*., 2000). Despite this observation, a comparison of samples collected during 1973–1974 and 1994–1997 indicated that streamwater NO_3^- concentrations have significantly decreased over this 20 yr period (Martin *et al*., 2000).

3.8.3. *Influence of Fire/Land-use*

While forest successional status and logging history are important, other land disturbances such as fires, agriculture and grazing, may strongly affect N retention. A good example of the influence of fire on N retention is at the Cone Pond watershed, which strongly retained N on an annual basis (96% DIN retention) and had outputs of NO_3-N that were among the lowest of the streams included in our study. The Cone Pond watershed is predominantly coniferous and is comprised of uneven-aged trees, some of which are over 250 yr old. Only a small proportion of the Cone Pond watershed has been harvested; however, approximately 85% of the watershed was heavily burned around 1820 as indicated by the presence of soil charcoal (Buso *et al*., 1984; Hornbeck and Lawrence, 1997).

In the few years following fire, streamwater DIN export may increase as a result of higher nitrification associated with warmer soil temperatures, and greater DIN runoff due to reduced evapotranspiration (Tiedemann *et al.*, 1978; Wright, 1976). However, this pulsed release of DIN to streams is generally short-lived, as DIN is rapidly taken up by aggrading vegetation (Bayley *et al.*, 1992; Bormann and Likens, 1979; Brown *et al.*, 1973; Schindler *et al.*, 1980). Long-term effects of severe fires typically reduce soil C and N storage by volatilization of C and N compounds. The fire at Cone Pond is thought to have been sufficiently severe to remove most of the soil organic matter, thereby reducing soil C and N content. The initial loss of C and N was followed by re-growth of red spruce (*Picea rubens*) and balsam fir (*Abies balsamea*) vegetation, which has poor quality litter with a high lignin:N ratio. Currently, soil C:N ratios in the burned areas of the watershed are high (>30:1) compared to unburned areas (17:1) (Hornbeck and Lawrence, 1997). High soil C:N ratios and poor litter quality may limit nitrification and NO_3-N production, causing a reduction in NO_3-N leaching. These findings suggest that although the fire at Cone Pond occurred over 180 yr ago, there has been a lasting effect on C and N pools resulting in low NO_3-N exports.

Data from the paired watershed study at Acadia reinforces our interpretation of the influence of fire on N retention. The Hadlock Brook watershed at Acadia has been left largely undisturbed, whereas the neighboring Cadillac Brook watershed was largely burned by wildfire in 1947. Although many of the characteristics between the two Acadia watersheds are similar, the DIN outputs are much lower at Cadillac Brook (0.2 kg N ha^{-1} yr^{-1}) compared to Hadlock Brook (1.3 kg N ha^{-1} yr^{-1}) (Nelson, 2002). At Leading Ridge, the upper portion of the watershed was clear-cut in the mid to late 1800's for charcoal production, and was severely burned during this period. The lower portion of the watershed was used as pastureland until the late 1890's. The Cockaponset watershed was also used as pastureland prior to re-growth of the present forest. All of these land-use practices may reduce the soil C and N stores resulting in low stream NO_3-N losses (<0.1 kg N ha^{-1} yr^{-1}) and high (nearly 100%) DIN retention.

4. Conclusions

Export of DIN in streamwater was less than wet-only DIN input at all of the watersheds included in our study. However, the large differences in percent N retention indicate that watersheds vary widely in their ability to retain N. Some watersheds retained nearly all of the wet N deposited on an annual basis, whereas other watersheds had outputs that were closer to wet N inputs. High streamwater exports of N may be an indication that some watersheds are approaching a condition of N saturation.

Data from Europe show that significant N leaching occurs when inputs exceed 25 kg N ha^{-1} yr^{-1}. In contrast, differences in N retention among watersheds in our study were not directly related to N loading. Rather these differences appear to be the result of a complex combination of factors involving vegetation, land-use, geology, and soils. These controls affect C and N pools within watersheds and ultimately influence the release of N to streams.

In recent years, data from watershed studies have provided advances in our understanding of N cycling in forested watersheds. However, many unanswered questions still remain, such as those related to the importance of hydrology, vegetation influences, disturbance, denitrification, N fixation, and dry N deposition. The role of some of these factors has been addressed at the plot or watershed level, but is still poorly understood on a regional scale. The results presented here suggest that regional analyses combined with specific case studies are needed to evaluate the spatial and temporal patterns of N solute loss in surface waters of the northeastern United States.

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References

Aber, J., McDowell, W., Nadelhoffer, K., Magill, A., Berntson, G., Kamakea, M., McNulty, S., Currie, W., Rustad, L. and Fernandez, I.: 1998, ‘Nitrogen saturation in temperate forest ecosystems’, *BioScience* **48**, 921–934.

Aber, J. D., Magill, A., McNulty, S. G., Boone, R. D., Nadelhoffer, K. J., Downs, M. and Hallett, R.: 1995, ‘Forest biogeochemistry and primary production altered by nitrogen saturation’, *Water, Air, and Soil Pollut.* **85**, 1665–1670.

Aber, J. D., Nadelhoffer, K. J., Steudler, P. and Melillo, J. M.: 1989, ‘Nitrogen saturation in northern forest ecosystems’, *BioScience* **39**, 378–386.

Aber, J. D., Ollinger, S. V., Driscoll, C. T., Likens, G. E., Holmes, R. T., Freuder, R. J. and Goodale, C. L.: 2002, 'Inorganic N losses from a forested ecosystem in response to physical, chemical, biotic and climatic perturbations', *Ecosystems* **5**, 648–658.

Adams, M. B., Angradi, T. R. and Kochenderfer, J. N.: 1997, 'Stream water and soil solution responses to 5 years of nitrogen and sulfur additions at the Fernow Experimental Forest, West Virginia', *For. Ecol. Manage.* **95**, 79–91.

Ågren, G. I. and Bosatta, E.: 1988, 'Nitrogen saturation of terrestrial ecosystems', *Environ. Pollut.* **54**, 185–197.

Ashby, J. A., Bowden, W. B. and Murdoch, P. S.: 1998, 'Controls on dentrification in riparian soils in headwater catchments of a hardwood forest in the Catskill Mountains, U.S.A.', *Soil Biol. Biochem.* **30**, 853–864.

Bayley, S. E., Schindler, D. W., Beaty, K. G., Parker, B. R. and Stainton, M. P.: 1992, 'Effects of multiple fires on nutrient yields from streams draining boreal forest and fen watersheds: Nitrogen and phosphorus', *Can. J. Fish. Aquat. Sci.* **49**, 584–596.

Bohlen, P. J., Groffman, P. M., Driscoll, C. T., Fahey, T. J. and Siccama, T. G.: 2000, 'Plant-soil-microbial interactions in a northern hardwood forest', *Ecology* **82**, 965–978.

Bormann, B. T., Bormann, F. H., Bowden, W. B., Pierce, R. S., Hamburg, S. P., Wang, D., Snyder, M. C. and Ingersoll, R. C.: 1993, 'Rapid N_2 fixation in pines, alder, and locust: Evidence from the sandbox ecosystem study', *Ecology* **74**, 583–598.

Bormann, F. H. and Likens, G. E.: 1979, *Pattern and Process in a Forested Ecosystem*, Springer-Verlag, New York, pp. 253.

Bowden, R. D., Steudler, P. A. and Melillo, J. M.: 1990, 'Annual nitrous oxide fluxes from temperate forest soils in the northeastern United States', *J. Geophys. Res.* **95**, 13997–14005.

Bowden, W. B.: 1986, 'Gaseous nitrogen emissions from undisturbed terrestrial ecosystems: An assessment of their impacts on local and global nitrogen budgets', *Biogeochemistry* **2**, 249–279.

Brown, G. W., Gahler, A. R. and Marston, R. B.: 1973, 'Nutrient losses after clear-cut logging and slash burning in the Oregon Coast Range', *Water Resour. Res.* **9**, 1450–1453.

Burns, D. A., Murdoch, P. S., Lawrence, G. B. and Michel, R. L.: 1998, 'Effect of groundwater springs on NO_3^- concentrations during summer in Catskill Mountain streams', *Water Resour. Res.* **34**, 1987–1996.

Buso, D. C., Martin, C. W. and Hornbeck, J. W.: 1984, 'Potential for Acidification of Six Remote Ponds in the White Mountains of New Hampshire', New Hampshire Water Resources Research Center, *Res. Rep. No. 43*.

Campbell, J. L., Eagar, C. and McDowell, W. H.: 2002, 'Patterns of streamwater acidity in the Lye Brook Wilderness Area, Vermont, U.S.A.', *Environ. Manage.* **30**, 234–248.

Campbell, J. L., Hornbeck, J. W., McDowell, W. H., Buso, D. C., Shanley, J. B. and Likens, G. E.: 2000, 'Dissolved organic nitrogen budgets for upland, forested ecosystems in New England', *Biogeochemistry* **49**, 123–142.

Castro, M. S. and Morgan, R. P.: 2000, 'Input-output budgets of major ions for a forested watershed in western Maryland', *Water, Air, and Soil Pollut.* **119**, 121–137.

Christ, M., Zhang, Y., Likens, G. E. and Driscoll, C. T.: 1995, 'Nitrogen retention capacity of a northern hardwood forest soil under ammonium sulfate additions', *Ecol. Appl.* **5**, 802–812.

Church, M. R.: 1997, 'Hydrochemistry of forested catchments', *Ann. Rev. Earth Planet. Sci.* **25**, 23–59.

Clarke, J. F., Edgerton, E. S. and Martin, B. E.: 1997, 'Dry deposition calculations for the clean air status trends network', *Atmos. Environ.* **31**, 3667–3678.

Creed, I. F. and Band, L. E.: 1998, 'Export of nitrogen from catchments within a temperate forest: Evidence for a unifying mechanism regulated by variable source area dynamics', *Water Resour. Res.* **34**, 3105–3120.

Dingman, S. L., Seely-Reynolds, D. M. and Reynolds, R. C.: 1988, 'Application of kriging to estimating mean annual precipitation in a region of orographic influence', *Water Resour. Bull.* **24**, 329–339.

Dise, N. B. and Wright, R. F.: 1995, 'Nitrogen leaching from European forests in relation to nitrogen deposition', *For. Ecol. Manage.* **71**, 153–161.

Driscoll, C. T., Lawrence, G. B., Bulger, A. J., Butler, T. J., Cronan, C. S., Eagar, C., Lambert, K. F., Likens, G. E., Stoddard, J. L. and Weathers, K. C.: 2001, 'Acidic deposition in the northeastern United States: Sources and inputs, ecosystem effects, and management strategies', *BioScience* **51**, 180–198.

Edwards, P. J. and Helvey, J. D.: 1991, 'Long-term ionic increases from a central Appalachian forested watershed', *J. Environ. Qual.* **20**, 250–255.

Federer, C. A.: 1997, 'BROOK90: A Simulation Model for Evaporation, Soil Water, and Streamflow, Version 3.2. Computer Freeware and Documentation', U.S. Department of Agriculture, Forest Service, Northeastern Forest Experiment Station, P.O. Box 640, Durham, NH 03824.

Federer, C. A. and Lash, D.: 1978, 'Simulated streamflow response to possible differences in transpiration among species of hardwood trees', *Water Resour. Res.* **14**, 1089–1097.

Galloway, J. N.: 1998, 'The global nitrogen cycle: Changes and consequences', *Environ. Pollut.* **102**, 15–24.

Galloway, J. N., Hendrey, G. R., Schofield, C. L., Peters, N. E. and Johannes, A. H.: 1987, 'Processes and causes of lake acidification during spring snowmelt in west-central Adirondack Mountains, New York', *Can. J. For. Res.* **44**, 1595–1602.

Gilliam, F. S., Adams, M. B. and Yurish, B. M.: 1996, 'Ecosystem nutrient responses to chronic nitrogen inputs at Fernow Experimental Forest, West Virginia', *Can. J. For. Res.* **26**, 196–205.

Gilliam, F. S., Yurish, B. M. and Adams, M. B.: 2001, 'Temporal and spatial variation of nitrogen transformations in nitrogen-saturated soils of a central Appalachian hardwood forest', *Can. J. For. Res.* **31**, 1768–1785.

Goodale, C. L., Aber, J. D. and McDowell, W. H.: 2000, 'The long-term effects of disturbance on organic and inorganic nitrogen export in the White Mountains, New Hampshire', *Ecosystems* **3**, 433–450.

Gundersen, P., Callesen, I. and deVries, W.: 1998, 'Nitrate leaching in forest ecosystems is related to forest floor C/N ratios', *Environ. Pollut.* **102**, 403–407.

Holloway, J. M. and Dahlgren, R. A.: 1999, 'Geologic nitrogen in terrestrial biogeochemical cycling', *Geology* **27**, 567–570.

Holloway, J. M., Dahlgren, R. A., Hansen, B. and Casey, W. H.: 1998, 'Contribution of bedrock nitrogen to high nitrate concentrations in stream water', *Nature* **395**, 785–788.

Hornbeck, J. W., Bailey, S. W., Buso, D. C. and Shanley, J. B.: 1997, 'Streamwater chemistry and nutrient budgets for forested watersheds in New England: variability and management implications', *For. Ecol. Manage.* **93**, 73–89.

Hornbeck, J. W. and Lawrence, G. B.: 1997, 'Eastern Forest Fires can have Long-term Impacts on Nitrogen Cycling', in *Diverse Forests, Abundant Opportunities, and Evolving Realities, Proceedings 1996 Society of American Foresters Convention, 9–13 November*, Albuquerque, NM, SAF Publ. 97-03, pp. 435–436.

Hornbeck, J. W., Smith, C. T., Martin, C. W., Tritton, L. M. and Pierce, R. S.: 1990, 'Effects of intensive harvesting on nutrient capitals of three forest types in New England', *For. Ecol. Manage.* **30**, 55–64.

Ito, M., Mitchell, M. J. and Dricsoll, C. T.: 2002, 'Spatial patterns of precipitation quantity and chemistry and air temperature in the Adirondack region of New York', *Atmos. Environ.* **36**, 1051–1062.

Kahl, J., Norton, S., Fernandez, I., Rustad, L. and Handley, M.: 1999, 'Nitrogen and sulfur input-output budgets in the experimental and reference watersheds, Bear Brook Watershed in Maine (BBWM)', *Environ. Monit. Assess.* **55**, 113–131.

Kahl, J. S., Norton, S. A., Fernandez, I. J., Nadelhoffer, K. J., Driscoll, C. T. and Aber, J. D.: 1993, 'Experimental inducement of nitrogen saturation at the watershed scale', *Environ. Sci. Technol.* **27**, 565–568.

Lajtha, K., Seely, B. and Valiela, I.: 1995, 'Retention and leaching losses of atmospherically-derived nitrogen in the aggrading coastal watershed of Waquoit Bay, MA', *Biogeochemistry* **28**, 33–54.

Lewis, G. P. and Likens, G. E.: 2000, 'Low stream nitrate concentrations associated with oak forests on the Allegheny High Plateau of Pennsylvania', *Water Resour. Res.* **36**, 3091–3094.

Likens, G. E. and Bormann, F. H.: 1995, *Biogeochemistry of a Forested Ecosystem*, Springer-Verlag, New York, pp. 159.

Lovett, G. M. and Kinsman, J. D.: 1990, 'Atmospheric pollutant deposition to high-elevation ecosystems', *Atmos. Environ.* **24**, 2767–2786.

Lovett, G. M. and Lindberg, S. E.: 1993, 'Atmospheric deposition and canopy interactions of nitrogen in forests', *Can. J. For. Res.* **23**, 1603–1616.

Lovett, G. M. and Rueth, H.: 1999, 'Soil nitrogen transformations in beech and maple stands along a nitrogen deposition gradient', *Ecol. Appl.* **9**, 1330–1344.

Lovett, G. M., Weathers, K. C. and Sobczak, W. V.: 2000, 'Nitrogen saturation and retention in forested watersheds of the Catskill Mountains, New York', *Ecol. Appl.* **10**, 73–84.

Lynch, J. A. and Corbett, E. S.: 1989, 'Hydrologic control of sulfate mobility in a forested watershed', *Water Resour. Res.* **25**, 1695–1703.

Magill, A. H., Aber, J. D., Berntson, G. M., McDowell, W. H., Nadelhoffer, K. J., Melillo, J. M. and Steudler, P.: 2000, 'Long-term nitrogen additions and nitrogen saturation in two temperate forests', *Ecosystems* **3**, 238–253.

Martin, C. W., Driscoll, C. T. and Fahey, T. J.: 2000, 'Changes in streamwater chemistry after 20 years from forested watersheds in New Hampshire, U.S.A.', *Can. J. For. Res.* **30**, 1–8.

McHale, M., Mitchell, M. J., McDonnell, J. J. and Cirmo, C. P.: 2000, 'Nitrogen solutes in an Adirondack forested watershed: Importance of dissolved organic nitrogen', *Biogeochemistry* **48**, 165–184.

McNulty, S. G., Aber, J. D. and Boone, R. D.: 1991, 'Spatial changes in forest floor and foliar chemistry of spruce-fir forests across New England', *Biogeochemistry* **14**, 13–29.

McNulty, S. G., Aber, J. D. and Newman, S. D.: 1996, 'Nitrogen saturation in a high elevation New England spruce-fir stand', *For. Ecol. Manage.* **84**, 109–121.

Miller, E. K., Friedland, A. J., Arons, E. A., Mohnen, V. A., Battles, J. J., Panek, J. A., Kadlecek, J. and Johnson, A. H.: 1993, 'Atmospheric deposition to forests along an elevational gradient at Whiteface Mountain, NY, U.S.A.', *Atmos. Environ.* **27A**, 2121–2136.

Mitchell, M. J.: 2001, 'Linkages of nitrate losses in watersheds to hydrological processes', *Hydrol. Process.* **15**, 3305–3307.

Mitchell, M. J., Driscoll, C. T., Kahl, J. S., Likens, G. E., Murdoch, P. S. and Pardo, L. H.: 1996, 'Climatic control of nitrate loss from forested watersheds in the northeast United States', *Environ. Sci. Technol.* **30**, 2609–2612.

Mitchell, M. J., Driscoll, C. T., Owen, J. S., Schaefer, D., Michener, R. and Raynal, D. J.: 2001a, 'Nitrogen biogeochemistry of three hardwood ecosystems in the Adirondack Mountains of New York', *Biogeochemistry* **56**, 93–133.

Mitchell, M. J., McHale, P. J., Inamdar, S. and Raynal, D. J.: 2001b, 'Role of within lake processes and hydrobiogeochemical changes over 16 yr in a watershed in the Adirondack Mountains of New York State, U.S.A.', *Hydrol. Process.* **15**, 1951–1965.

Munger, J. W. and Eisenreich, S. J.: 1983, 'Continental-scale variations in precipitation chemistry', *Environ. Sci. Technol.* **17**, 32A–42A.

Murdoch, P. S., Burns, D. A. and Lawrence, G. B.: 1998, 'Relation of climate change to the acidification of surface waters by nitrogen deposition', *Environ. Sci. Technol.* **32**, 1642–1647.

Murdoch, P. S. and Stoddard, J. L.: 1992, 'The role of nitrate in the acidification of streams in the Catskill Mountains of New York', *Water Resour. Res.* **28**, 2707–2720.

Nadelhoffer, K. J., Downs, M. R., Fry, B., Aber, J. D., Magill, A. H. and Melillo, J. M.: 1995, 'The fate of ^{15}N-labelled nitrate additions to a northern hardwood forest in eastern Maine, U.S.A.', *Oecologia* **103**, 292–301.

NADP: 2002, National Atmospheric Deposition Program (NRSP-3)/National Trends Network. NADP Program Office, Illinois State Water Survey, 2204 Griffith Drive, Champaign, IL 61820.

Nelson, S. J.: 2002, 'Determining Atmospheric Inputs to Two Small Watersheds at Acadia National Park', *M.S. Thesis*, University of Maine, pp. 112.

Norton, S. A. and Fernandez, I. J.: 1999, *The Bear Brook Watershed in Maine: A Paired Watershed Experiment – The First Decade (1987–1997)*, Kluwer Academic Publishers, Boston, pp 250.

Ollinger, S. V., Aber, J. D., Federer, C. A., Lovett, G. M. and Ellis, J. M.: 1995, 'Modeling physical and chemical climate of the northeastern United States for a geographic information system', *Gen. Tech. Rep. NE-191*. Radnor, PA: U.S. Department of Agriculture, Forest Service, Northeastern Forest Experiment Station, p. 30.

Peterjohn, W. T., Adams, M. B. and Gilliam, F. S.: 1996, 'Symptoms of nitrogen saturation in two central Appalachian hardwood forest ecosystems', *Biogeochemistry* **35**, 507–522.

Peterjohn, W. T., Foster, C. J., Christ, M. J. and Adams, M. B.: 1999, 'Patterns of nitrogen availability within a forested watershed exhibiting symptoms of nitrogen saturation', *For. Ecol. Manage.* **119**, 247–257.

Peterjohn, W. T., McGervey, R. J., Sexstone, A. J., Christ, M. J., Foster, C. J. and Adams, M. B.: 1998, 'Nitrous oxide production in two forested watersheds exhibiting symptoms of nitrogen saturation', *Can. J. For. Res.* **28**, 1723–1732.

Rascher, C. M., Driscoll, C. T. and Peters, N. E.: 1987, 'Concentration and flux of solutes from snow and forest floor during snowmelt in the West-Central Adirondak region of New York', *Biogeochemistry* **3**, 209–224.

Schindler, D. W., Newbury, R. W., Beaty, K. G., Prokopowich, J., Ruszczynski, T. and Dalton, J. A.: 1980, 'Effects of a windstorm and forest fire on chemical losses from forested watersheds and on the quality of receiving streams', *Can. J. Fish. Aquat. Sci.* **37**, 328–334.

Skeffington, R. A. and Wilson, E. J.: 1988, 'Excess nitrogen deposition: Issues for consideration', *Environ. Pollut.* **54**, 159–184.

Stoddard, J. L.: 1994, 'Long-term Changes in Watershed Retention of Nitrogen', in L. A. Baker (ed.), *Environmental Chemistry of Lakes and Reservoirs*, American Chemical Society, Washington, D.C., pp. 223–284.

Swistock, B. R., Edwards, P. J., Wood, F. and DeWalle, D. R.: 1997, 'Comparison of methods for calculating annual solute exports from six forested Appalachian watersheds', *Hydrol. Process.* **11**, 655–669.

Tiedemann, A. R., Helvey, J. D. and Anderson, T. D.: 1978, 'Stream chemistry and watershed nutrient economy following wildfire and fertilization in eastern Washington', *J. Environ. Qual.* **7**, 580–588.

Vitousek, P. M. and Reiners, W. A.: 1975, 'Ecosystem succession and nutrient retention: A hypothesis', *BioScience* **25**, 376–381.

Weathers, K. C., Likens, G. E., Bormann, F. H., Bicknell, S. H., Bormann, B. T., Daube, B. C., Eaton, J. S., Galloway, J. N., Keene, W. C., Kimball, K. D., McDowell, W. H., Siccama, T. G., Smiley, D. and Tarrant, R. A.: 1988, 'Cloudwater chemistry from ten sites in North America', *Environ. Sci. Technol.* **22**, 1018–1026.

Weathers, K. C., Lovett, G. M., Likens, G. E. and Lathrop, R.: 2000, 'The effect of landscape features on deposition to Hunter Mountain, Catskill Mountains, New York', *Ecol. Appl.* **10**, 528–540.

Wigington, P. J., DeWalle, D. R., Murdoch, P. S., Kretser, W. A., Simonin, H. A., Van Sickle, J. and Baker, J. P.: 1996, 'Episodic acidification of small streams in the northeastern United States: Ionic controls of episodes', *Ecol. Appl.* **6**, 389–407.

Wright, R. F.: 1976, 'The impact of forest fire on the nutrient influxes to small lakes in northeastern Minnesota', *Ecology* **57**, 649–663.